A colored-noise Fokker-Planck equation for non-Brownian particles in shear-induced diffusion

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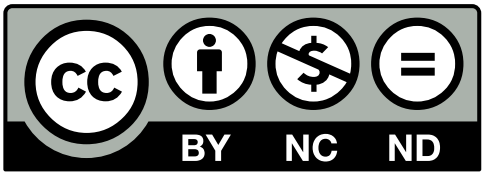

Abstract

The topic of the present work are non-Brownian particles in shear flow. As reported in literature, the occurring phenomenon in this context is shear-induced diffusion which takes place in the absence of the well-known Brownian diffusion. Diffusive processes can be described stochastically in terms of a stochastic differential equation (Langevin equation or Langevin-like equation) or a differential equation for the probability density, in second order referred to as Fokker-Planck equation. It is known that in contrast to Brownian diffusion, the shear-induced diffusion is a long-time diffusion which poses a challenge to the stochastic description of this phenomenon. The present work analyzes the problem of non-Brownian particles in shear-induced diffusion with regard to the Markov property of the treated variables. This concludes that the Fokker-Planck equation so far derived in pure position space may not be sufficient. In order to ensure the Markov process property, a Fokker-Planck equation extended to coupled position colored-noise velocity space is derived. Throughout the extension, the colored-noise velocity is modeled as an Ornstein-Uhlenbeck process. These first two steps were also treated in the author's Master thesis (Lukassen 2012). A detailed substantiation of this approach is published in (Lukassen & Oberlack 2014b) including a new multiple time scale analysis and a Gaussian solution. The multiple time scale analysis results in the dimensionless form of the equation of motion which serves as a starting point for the derivation of the new colored-noise Fokker-Planck equation. As a next step, this coupled Fokker-Planck equation is integrated over velocity space and approximated to yield a reduced position-space Fokker-Planck equation. It is shown that such a reduction as in the present work is only possible under certain conditions concerning the correlation time. The resulting position-space equation is analyzed and compared to the traditional position-space models. The reduced form exhibits additional correction terms. In an outlook, possible extensions of the presented model are discussed with exemplary simulation results. Chapter 5 and 6 as a whole are based on the author's publication (Lukassen & Oberlack 2014b).

Zusammenfassung

Das Thema der vorliegenden Arbeit sind nicht-Brownsche Partikel in Scherströmung. Obwohl nicht-Brownsche Partikel keine Brownsche Molekularbewegung ausführen, ist aus der Literatur bekannt, dass in diesem Zusammenhang das Phänomen der scher-induzierten Diffusion auftritt. Diffusive Prozesse lassen sich stochastisch beschreiben. Dazu kann eine Langevin Gleichung, beziehungsweise eine Langevin-ähnliche Gleichung herangezogen werden. Eine andere Möglichkeit besteht darin, die Differentialgleichung der Wahrscheinlichkeitsdichte zu benutzen, die in zweiter Ordnung Fokker-Planck Gleichung genannt wird. Es ist bekannt, dass die scher-induzierte Diffusion, im Gegensatz zur Brownschen Diffusion, eine Langzeit-Diffusion ist, was die stochastische Beschreibung erschwert. In dieser Arbeit werden nicht-Brownsche Partikel in Scherströmung mit Hinblick auf die Markov Eigenschaft der zugrunde liegenden Variablen untersucht. Daraus resultiert die Annahme, dass die Fokker-Planck Gleichung, die bisher zur Beschreibung nicht-Brownscher Partikel genutzt wurde, eine Gleichung nur in Ortsvariablen, nicht ausreichend sein könnte. Um die Markov Eigenschaft zu gewährleisten, wird die Fokker-Planck Gleichung hier um den Geschwindigkeitsraum erweitert, wobei die hydrodynamische Geschwindigkeitskomponente als farbiges Rauschen modelliert wird. Basis für das hier aufgestellte Modell ist ein Ornstein-Uhlenbeck Prozess. Diese beiden ersten Schritte sind auch in der Master Arbeit (Lukassen 2012) behandelt worden. Eine substantielle Herleitung wurde in (Lukassen & Oberlack 2014b) veröffentlicht. Darin enthalten ist eine neue Zeitskalenanalyse und eine Gaußsche Lösung der neuen Fokker-Planck Gleichung. Desweiteren wird die gekoppelte Fokker-Planck Gleichung über den Geschwindigkeitsraum integriert und im zweiten Schritt approximiert und die reduzierte Ortsraum Gleichung mit den bisher verwendeten Modellen im Ortsraum verglichen. Diese neue Formulierung weist zusätzliche Korrekturterme auf. Bei einer solchen Reduzierung müssen einschränkende Bedingungen an die Korrelationszeit spezifiziert werden. In einem Ausblick werden mögliche Erweiterungen des vorgestellten Modells erläutert und an exemplarischen Simulationsergebnissen diskutiert. Die Kapitel 5 und 6 basieren auf der Veröffentlichung (Lukassen & Oberlack 2014b).

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Nomenclature

α	The regarded particle
$\boldsymbol{\Omega}$	Particle angular velocity
$\boldsymbol{D}$	Diffusion matrix
$\boldsymbol{E}^{\infty}$	Rate of strain
$\boldsymbol{F}(\boldsymbol{x})$	Position-dependent external force field
$\boldsymbol{F}^{B}$	Brownian force
$\boldsymbol{F}^{H}$	Hydrodynamic force
$\boldsymbol{F}^{P}$	Interparticle or external force
$\boldsymbol{F}^{bass}$	Basset history force
$\boldsymbol{f}^{ext}(\boldsymbol{x})$	External force per unit volume
$\boldsymbol{F}_{\alpha}^{op}$	Hydrodynamic force onto particle α due to surrounding particles
$\boldsymbol{F}^{vm}$	Virtual mass force
$\boldsymbol{I}$	Unity matrix
$\boldsymbol{R}_{FE}$	Resistance matrices relating force and rate of strain
$\boldsymbol{R}_{FU}$	Resistance matrices relating force and velocity
$\boldsymbol{U}$	Particle velocity
$\boldsymbol{u}(\boldsymbol{x},t)$	Fluid velocity at position $\boldsymbol{x}$
$\boldsymbol{U}^{\infty}$	Shear velocity
$\boldsymbol{u}^{fluid}(\boldsymbol{x},t)$	Undisturbed fluid velocity
$\boldsymbol{V}_{\alpha}$	Colored-noise velocity of particle α
$\boldsymbol{X}$	Stochastic multidimensional variable
$\delta(t-t')$	Dirac impulse
$\dot{\gamma}$	Shear rate
ϵ_1, ϵ_2	Expansion parameter for time scale analysis
η	Viscosity of the solvent
η_r	Relative viscosity
η_{eff}	Effective viscosity
κ_m	Cumulants in one dimension

$\langle\langle\rangle\rangle$ Denotes multidimensional cumulants

$\mathbb{P}$ Probability distribution function

μ_m m-th moment

ϕ Particle volume fraction

ϕ_{max} Maximal particle volume fraction

$\Phi_{pot}(r)$ Interaction potential

ψ Noise term

ρ_f Fluid density

ρ_p Particle density

τ Time scale

τ_a Placeholder time scale for the regarded time scale

τ_c Correlation time for colored-noise velocity

τ_D Diffusive time scale for Brownian particles

τ_f Shear flow time scale

τ_p Inertial relaxation time scale

τ_{ac} Placeholder time scale for the velocity autocorrelation

τ_{conf} Placeholder time scale for change of particle configuration

τ_{fluid} Fluid time scale

τ_{mol} Time scale of molecules

$\triangle t$ Time step

ξ White noise term

a Particle radius

$C(u)$ Characteristic function

D_0 Stokes-Einstein diffusivity

D_{ij} Diffusion coefficients

$fr = 6\pi\eta a$ Stokes friction coefficient

k Boltzmann constant

L White noise in velocity model

m Mass of a particle

P Probability distribution

$p(\boldsymbol{x}, t)$ Pressure

$P_r(x_1, \ldots, x_r)$ Multivariate probability distribution

Pe Péclet number

Re_p	particle Reynolds number
Re_U	Reynolds number
St	Stokes number
T	Absolute temperature
t	Time
U	Particle velocity component
$Y_x(t)$	Variable in a stochastic process

1 Introduction

The scope of the present work are non-Brownian solid particles in shear flow. Even though the name non-Brownian implies the absence of the well-known Brownian diffusion, these particles perform diffusive motion. This phenomenon is called shear-induced diffusion, which results purely from the hydrodynamic interaction between the particles. In this way it differs from the Brownian diffusion and turbulent diffusion (Breedveld, van den Ende, Bosscher, Jongschaap & Mellema 2002). Shear-induced diffusion has first been found experimentally by e.g. Eckstein, Bailey & Shapiro (1977) and Breedveld, van den Ende, Tripathi & Acrivos (1998). Also, in numerical simulations this diffusive behavior could be substantiated, e.g. in (Sierou & Brady 2004). It was found that indeed enough many-particle hydrodynamic interactions suffice to create diffusive behavior (Sierou & Brady 2004). Such a stochastic process can be described by the equation of motion in form of a stochastic differential equation, referred to as Langevin equation, and equivalently by the Fokker-Planck equation, a differential equation for the probability density, cf. e.g. (van Kampen 2007).

Brownian particles exhibit a so-called separation of time scales in the sense that their velocity is correlated on a much smaller time scale than the position changes (Dhont 1996). This enables to describe the Brownian position and velocity in two separate equations, see (van Kampen 2007). In contrast to that, non-Brownian particles do not exhibit separate time scales. The only time scale inherent in the system is given by the shear rate, so the configuration and velocity change on the same time scale and the diffusion is a so-called long-time diffusion, see (Sierou & Brady 2004). The non-separation of time scales poses difficulties in deriving the appropriate statistical equations to describe non-Brownian particles in shear flow. The non-Brownian Langevin equation has been described in an N-particle form by Breedveld et al. (2002) assuming that the influence of the particles onto each other can be modeled as a colored-noise force. The Fokker-Planck equation for shear-induced diffusion has, so far, only been derived in pure position space, (Sierou & Brady 2004), (Breedveld et al. 1998), (Santamaría-Holek, Barrios & Rubi 2009b).

As will be shown throughout the present thesis, the pure position-space description may not be sufficient to describe the phenomenon of shear-induced behavior of non-Brownian particles and an alternative Fokker-Planck equation is derived. A new multiple time scale analysis helps to substantiate this new approach. Further, the resultant colored-noise Fokker-Planck equation is solved. For that, a Gaussian approach is used as given in (van Kampen 2007).
The colored-noise Fokker-Planck equation is furthermore reduced to a new form in position space employing an asymptotic assumption. Here, the approaches of Wilemski (1976) and Brinkman (1956), Sack (1956) are combined. The resultant reduced

position-space equation equals in lowest order the traditional position-space equations mentioned above but exhibits higher-order correction terms.

1.1 Outline of the present work

The present work introduces a framework for the description of non-Brownian particles in shear-induced diffusion. In chapter 2 a general introduction to particle flows is given. Most importantly, the difference between Brownian and non-Brownian particles is outlined in section 2.3. This contains a description of shear-induced diffusion in comparison to Brownian diffusion. Chapter 3 treats the equation of motion of particles in the form of a stochastic differential equation. The mathematical substantiation of such an equation is summarized from the literature. Further, the Langevin equation as originally derived, namely for a single Brownian particle, is given from (Risken 1989) (section 3.1). As reported in the literature, the original form of the Langevin equation gives rise to criticism, see section 3.1.3.1, which will be referred to in chapter 8 again. As outlined above, shear-induced diffusion arises through the hydrodynamic interaction of many particles. In this sense, section 3.2 is essential for the present work as it introduces the Langevin equation for many hydrodynamically interacting particles based on Brady & Bossis (1988). Before proceeding with the Langevin equation for many particles, chapter 4 gives an introduction to probability theory. In section 4.2, an introduction to the Fokker-Planck equation is given. Special attention is paid to Markov processes in section 4.2.1 and the Ornstein-Uhlenbeck process 4.2.3.2. A Fokker-Planck equation can be set up on the time scale of interest. In this sense, it corresponds to a Langevin equation referring to the very same time scale, see e.g. (van Kampen 2007), (Dhont 1996). In section 4.3, the Fokker-Planck equation for Brownian particles is outlined in the context of different time scales (van Kampen 2007). In chapter 5, a new multiple time scale approach is given where the Langevin equation for many hydrodynamically interacting particles, introduced in chapter 3.2 from (Brady & Bossis 1988), is analyzed with regard to the Markov property of the position and velocity variable. The result is that only a coupled variable of position and velocity fulfills the Markov process assumption. Chapter 5 is heavily based on (Lukassen & Oberlack 2014b). Breedveld et al. (2002) introduce the assumption that the hydrodynamic force onto non-Brownian particles is colored-noise correlated. This assumption is adopted and a one-particle equation with a colored-noise velocity is formulated. In chapter 6, a new Fokker-Planck equation in coupled position colored-noise velocity space is derived ensuring the Markov property. This derivation includes a model for the hydrodynamic velocity component where an Ornstein-Uhlenbeck process is used. The resultant equation can be solved by a Gaussian approach following (van Kampen 2007). The new Fokker-Planck equation presented in chapter 6 has first been shown in (Lukassen 2012), chapter 6 as a whole is heavily based on (Lukassen & Oberlack 2014b) where a fundamental derivation is given. In this sense, the argumentation throughout this work towards the new formulation also follows these references. The resultant position colored-noise Fokker-Planck equation is reduced to a new form in position space combining the approaches of Wilemski (1976) and Brinkman (1956), Sack (1956),

Risken (1989) in chapter 7. Wilemski's approach implies the derivation of higher moment equations for the velocity in position space. The approach of Brinkman (1956), Sack (1956), Risken (1989) results in second order time Fokker-Planck-like equations. As a result, the new reduced position Fokker-Planck equation in chapter 7 is a second order in time equation. When comparing this to the traditional position-space equations used so far for non-Brownian particles in shear flow, it will be pointed out that the new reduced form contains additional correction terms. As one essential part of a detailed outlook in chapter 8, the criticism on the Langevin equation, outlined in section 3.1.3.1, is reconsidered for the present case and possible extensions of the presented model are proposed. This is substantiated by exemplary simulation results (for the used code see (Sierakowski & Prosperetti status January 23rd 2015)) gained in the scope of the present work.

2 Particle flows

The present work treats monodisperse non-Brownian spherical solid particles of negligible inertia suspended in a liquid solvent. The importance of particle inertia forces in relation to viscous fluid forces is given by the so-called Stokes number whereby negligible inertia means a Stokes number tending to zero, cf. (Mewis & Wagner 2013). Particles in shear flow are referred to as non-Brownian when the imposed shear flow dominates over Brownian motion, see e.g. (Foss & Brady 1999), (Foss & Brady 2000). Concerning size, particles in the range of colloids can perform Brownian motion, cf. (Dhont 1996). Non-colloidal particles are too large to perform Brownian motion and thus, are non-Brownian, cf. (Breedveld et al. 1998). Shear-induced diffusion has been observed for colloidal sized particles (Foss & Brady 1999) and for particle sizes up to $90\mu m$ which are too large for Brownian motion (Breedveld et al. 1998). The particles regarded in the present work shall be neutrally buoyant, thus with the particle density $\rho_p \approx \rho_f$, with ρ_f as fluid density. There shall be no effects such as sedimentation. The underlying flow behavior shall be a homogeneous laminar unbounded shear flow. Any wall interaction is avoided. The suspension medium shall be Newtonian. For the shear flow applies for the three velocity components in x, y, z-direction:

$$\boldsymbol{U}^{\infty} = (\dot{\gamma}y, 0, 0), \tag{2.1}$$

with the shear rate $\dot{\gamma}$. Bold symbols denote vectors, matrices and tensors throughout the present work. The scope of the present work is Stokes flow, which means that the inertia and acceleration terms in the Navier-Stokes equation are omitted (Mewis & Wagner 2013).

The particles in the present work shall be interacting purely hydrodynamically and the particle volume fraction shall be such that a lot of many-body interactions occur (for values of such volume fractions refer to (Sierou & Brady 2004)) which results in a diffusive behavior, namely shear-induced diffusion. A literature overview over particle flows in general will be given in section 2.1. Constitutive equations for the fluid and the forces onto particles are subject of section 2.2. Further, Brownian and non-Brownian particles are compared in section 2.3, in particular with respect to Brownian diffusion and shear-induced diffusion. In this context the investigation of several time scales is important. The chapter closes with a short note on viscosity in section 2.4.

2.1 Particle flows: General classification

In order to specify the characteristics of the particles that are regarded in the present work, here are shortly summarized important technical terms from literature. In

principle, a particle is defined as any condensed phase discontinuity in three dimensions, which can be solid, liquid or gaseous (Hackley & Ferraris 2001). In further specification, these could be grains (solid), droplets (liquid) or bubbles (gaseous) (Bernhardt 1994) or even aggregates (particles that form a cohesive mass or cluster) (Hackley & Ferraris 2001). Aggregates can be further classified with regard to the forces or ingredients that lead to the forming of the aggregate. One can also define gels, which may consist of particles or polymers combined with liquid (Hackley & Ferraris 2001). These discontinuities respectively particles build the dispersed phase in a second immiscible continuous phase (Hackley & Ferraris 2001). This two-phase system is called a dispersion (Hackley & Ferraris 2001). The continuous phase can also be solid, liquid or gaseous (Bernhardt 1994).
A liquid continuous phase with solid particles in it is considered in the present work and is called a suspension (Hackley & Ferraris 2001). This is the definition referred to throughout the present work. Still, in the literature there can be found more differentiated definitions: Mewis & Wagner (2013) state that the term suspension is rather used for particles of size larger than colloidal size (for size ranges of colloids, see below). A colloid is the dispersed phase in the two-phase system with specific size ranges (Mewis & Wagner 2013). For colloidal size particles in a suspending medium Mewis and Wagner use the term dispersion, Everett (1972) uses the term colloidal suspension. For the purpose of completeness, an emulsion would be droplets in a liquid continuous phase and gaseous particles in liquid would be foam or bubble systems (Bernhardt 1994). For more information, see (Schramm 2006).

The more general classification in this context is multiphase flow. Crowe, Schwarzkopf, Sommerfeld & Tsuji (2011) state that multiphase flows are either gas-liquid (e.g. bubbly flows, gas-droplet flows), gas-solid (e.g. fluidized beds known from industry where a gas suspends a bed of particles, gas-particle flows), liquid-solid (e.g. slurry flows, solid particles in liquid) or three-phase flows (e.g. bubbles in a slurry flow). Crowe et al. (2011) outline that the case of a fluid around motionless particles is commonly not referred to as liquid-solid or gas-solid flow but rather known as flow through a porous medium. This thesis focuses on liquid-solid flows, thus suspensions of solid particles in a liquid fluid.

2.1.1 Properties of solid particles and the solvent

After the definition and the context of the technical terms of suspensions of solid particles, the properties for particles and the continuous phase will be considered in the following. Nijenhuis, McKinley, Spiegelberg, Barnes, Aksel, Heymann & Odell (2007) modified a list of properties of the disperse and continuous phase which was in original given by Chander (1998). The following properties from their list are specified below: Particle size and size distribution, particle shape, and particle volume concentration. The latter is referred to as particle volume fraction (Crowe et al. 2011) in the present work.

Concerning particle sizes, Hackley & Ferraris (2001) give a very detailed classification which is summarized here. Molecules range up to $1nm$ (Nijenhuis et al. 2007), and will not be regarded in this work. Colloidal particles have a dimension between $1nm$ and $1\mu m = 1000nm$. This is the size range where particles may react to thermal fluctuations in the fluid (Brownian motion) (Dhont 1996), see also section 2.3.1. The upper limit of $1\mu m$ is not sharp, in (Dhont 1996) and (Russel, Saville & Schowalter 1991) is given a maximum size for colloids of $10\mu m$. Schramm (2006) states that the upper size limit of $1\mu m$ for colloidal particles is referred to spherical particles, whereby in the case of other shapes ranges to $2\mu m$ may also be described as colloids (in practical applications even up to $100\mu m$). More details about colloids can also be found in (Everett 1972). As of a size of a few micrometers the non-colloidal regime starts (Mewis & Wagner 2013), (Dhont 1996), (Russel et al. 1991). Proceeding in the classification according to Hackley & Ferraris (2001), ultrafine particles have a size range between $1\mu m$ and $10\mu m$. Fine refers to particles with maximal dimensions of $37\mu m$ and in case of at least one dimension exceeding $37\mu m$ the particle is coarse, followed by granule. The present work comprises non-Brownian particles in shear flow. This includes colloids where a shear flow dominates over the Brownian motion and non-colloids in the range of micrometers.

Further, one can distinguish between particulates where all particles have the same size (monodisperse) or not (heterodisperse), which for few different particle sizes would be paucidisperse and for many different particle sizes polydisperse (Everett 1972).

The shape of particles plays an important role. For example, in (Nijenhuis et al. 2007) it is stated that rod-formed particles may align in the flow and thus substantially influence the rheological behavior of the suspension. Non-spherical Brownian particles may also show rotational Brownian motion in addition to translational Brownian motion (Nijenhuis et al. 2007). Dhont (1996) examines the influence of rod particles onto the motion of the particles and states that in contrast to spherical particles, the translational motion of rod particles is coupled to the rotational motion of the particles. Throughout this work, the particles are of spherical shape which means that such coupled influences do not appear.

Additionally, particle flows can be classified as dilute, semi-dilute, and dense, cf. (Crowe et al. 2011), (Mewis & Wagner 2013). Crowe et al. (2011) give a broad introduction to the effects of dilute and dense flows which is summarized as follows: In case of dilute particle flow, the particle motion is mainly determined by fluid forces such as drag and lift; in dense flow by particle-particle interactions, cf. section 2.2.1. Crowe et al. (2011) differentiate between dilute and dense in terms of a ratio between momentum response times and times between particle-particle collisions. See also (Tadros 1996) for more information. The time scale of collisions as given in (Crowe et al. 2011) depends on the so-called particle volume fraction. The particle volume fraction can be used directly as a mean of measurement in that context, cf. (Mewis & Wagner 2013). The

particle volume fraction, also referred to as solid volume concentration (Nijenhuis et al. 2007) is defined as:

$$\phi = \frac{\text{volume of the dispersed phase}}{\text{volume of dispersed phase} + \text{volume of fluid phase}}. \tag{2.2}$$

For example a particle volume fraction up to 0.05 could be classified as dilute, followed by semi-dilute suspensions for $\phi \leq 0.15$ and then dense (Mewis & Wagner 2013). The maximum packing fraction ϕ_{max} is subject to various studies, in (Mewis & Wagner 2013) is given a broad literature overview on the derivations of values for ϕ_{max}. In this thesis, the particle volume fraction is assumed to be large enough so that enough many-body interactions occur for shear-induced diffusion to arise. Appropriate values can be found e.g. in (Sierou & Brady 2004). Sierou & Brady (2004) investigate shear-induced diffusion at various particle volume fractions which are also influenced by the number of particles starting at $\phi = 0.1$ for 1000 particles.

For the properties of the continuous phase, the listing from (Nijenhuis et al. 2007) and (Chander 1998) is used. More detailed information can be found there. For particles much larger than the fluid molecules, the fluid is considered as a continuum (Dhont 1996), (Russel et al. 1991). The continuous phase has a solvent viscosity η. Here, an aqueous Newtonian solvent without any other dissolved substances besides the regarded particles is considered. Consequently, all non-Newtonian effects that may arise come from the particle phase. For the influence of particles onto the effective viscosity of the suspension and possible non-Newtonian behavior, see section 2.4 and the sources given in there.

2.1.2 Definition: Time scales and time steps

The present work relies on an accurate definition of the terms time, time scale, and time step which will be introduced in this section. The following definitions can also be found in the author's publication (Lukassen & Oberlack 2014b). The time t is the independent variable to determine the generic range on which the process may vary, thus $t \in [t_0, \infty)$. A time scale τ defines the length of a process on which a significant change is observed, e.g. change in particle configuration, the period of a cyclic process or the relaxation time scale of an exponentially decaying event. Finally, a time step $\triangle t$ is a unit of the contemplated time scale, e.g. $\triangle t = \tau/q$ with $q \in \mathbb{N}$ (unless otherwise specified). A time step $\triangle t$ can also be a multiple of the contemplated time scale τ but it is important that $\triangle t$ is of the same order of magnitude as τ. So $\triangle t$ is inherently coupled to τ. For the time scales to appear in the present work, $\triangle t$ has to be chosen accordingly such that even in the limiting process of $\triangle t \to 0$, it is a given fraction of this time scale. That means, a time step $\triangle t \to 0$ with respect to a certain time scale will still be larger than the next smaller time scale (cf. (Dhont 1996)). In the present work, a $\triangle t \ll$ or $\gg \tau$ refers to a $\triangle t$ on the next smaller or larger time scale, whereby a $\triangle t <$ or $> \tau$ indicates a time step on the very same time scale τ.

2.2 Constitutive equations for the solvent and the particles

An important dimensionless number throughout the present work is the particle Reynolds number, cf. (Nijenhuis et al. 2007), (Mewis & Wagner 2013), given as (Dhont 1996):

$$\text{for the determining particle velocity component } U\text{: } Re_U = \frac{\rho_f a U}{\eta}, \tag{2.3}$$

$$\text{in shear flow: } Re_p = \frac{\rho_f a^2 \dot{\gamma}}{\eta}, \tag{2.4}$$

with ρ_f as fluid density, a particle radius, $\dot{\gamma}$ shear rate, and η as suspending medium viscosity. Note, that η is not the so-called effective medium viscosity called η_{eff}, see subsection 2.4 and the sources given in there. The particle Reynolds number gives the inertia of the fluid and is determined by the ratio of the inertial to viscous terms. An incompressible Newtonian fluid is determined by the Navier-Stokes equation with the continuity equation, cf. (Dhont 1996), (Russel et al. 1991):

$$\rho_f \left(\frac{\partial \boldsymbol{u}(\boldsymbol{x},t)}{\partial t} + \boldsymbol{u}(\boldsymbol{x},t) \cdot \nabla \boldsymbol{u}(\boldsymbol{x},t) \right) = \eta \nabla^2 \boldsymbol{u}(\boldsymbol{x},t) - \nabla p(\boldsymbol{x},t) + \boldsymbol{f}^{ext}(\boldsymbol{x}), \tag{2.5}$$

$$\nabla \cdot \boldsymbol{u}(\boldsymbol{x},t) = 0. \tag{2.6}$$

$\boldsymbol{u}(\boldsymbol{x},t)$ is the velocity of an infinitesimally small volume element at position $\boldsymbol{x}$ or just the local fluid velocity. $p(\boldsymbol{x},t)$ is the pressure. External fields may also induce forces onto the fluid which enter the equation as an external force per unit volume, $\boldsymbol{f}^{ext}(\boldsymbol{x})$. With that, the boundary conditions for the Navier-Stokes equation 2.5 are, (Maxey & Riley 1983):

$$\boldsymbol{u}(\boldsymbol{x},t) = \boldsymbol{U}(t) + \boldsymbol{\Omega}(t) \times (\boldsymbol{x} - \boldsymbol{x}_0) \text{ on the sphere} \tag{2.7}$$

$$\boldsymbol{u}(\boldsymbol{x},t) = \boldsymbol{u}^{fluid}(\boldsymbol{x},t) \text{ for } |\boldsymbol{x} - \boldsymbol{x}_0| \to \infty. \tag{2.8}$$

$\boldsymbol{u}^{fluid}(\boldsymbol{x},t)$ is the undisturbed fluid velocity in the absence of any particles. $\boldsymbol{x}_0$ is the location of the particle center. $\boldsymbol{U}$ is the particle velocity, $\boldsymbol{\Omega}$ is the angular velocity of the particle. The condition in equation 2.7 is the no-slip condition.
For the steady case, the time derivative is zero (Landau & Lifshitz 1963). Close to the particle surface, the velocity $\boldsymbol{u}$ scales as the particle velocity $\boldsymbol{U}$ (Dhont 1996). When lengths scale as the particle radius a in proximity to the particle (in the order of lengths comparable to the radius) then the gradient ∇ scales as $1/a$, see (Dhont 1996). Multiplying the whole equation 2.5 with $a^2 U/\eta$ (U as the order of the particle velocity) shows that the convective term is multiplied by Re_U (equation 2.3) and vanishes for $Re_U \to 0$. In general, the time derivative does not automatically vanish. It is important how the time derivative scales in comparison to U, η, a (Dhont 1996). In case that the regarded time scale is $\tau_f = 1/\dot{\gamma}$ and the reference particle velocity is $U = a\dot{\gamma}$, both sides of equation 2.5 may be multiplied by $a^2\dot{\gamma}/\eta$ which shows that both, the time derivative

term and the convective term are multiplied by Re_p and thus vanish for $Re_p \to 0$, see e.g. (Russel et al. 1991), (Dhont 1996), (Maxey & Riley 1983). In the present work, it is assumed that $Re_p \ll 1$. That means the local flow on the time scale τ_f and on the length scale of the particle is Stokes flow, also referred to as creeping flow conditions (Dhont 1996), (Mewis & Wagner 2013).

The Stokes drag (section 2.2.1) and the hydrodynamic interaction matrices introduced in section 3.2 are calculated on the basis of Stokes equation with small particle Reynolds number, see (Dhont 1996), (Durlofsky, Brady & Bossis 1987), (Brady & Bossis 1988). At larger distances from the particle, the nonlinear term in equation 2.5 is not zero anymore because the flow velocity $\boldsymbol{u}$ then scales as the undisturbed flow velocity $\boldsymbol{u}^{fluid}$ and the gradient $\nabla \boldsymbol{u}$ scales as the distance (Landau & Lifshitz 1963, p.61-68).

For the creeping flow conditions the Navier-Stokes equation 2.5 reduces to (Dhont 1996):

$$\nabla p(\boldsymbol{r}, t) - \eta \nabla^2 \boldsymbol{u}(\boldsymbol{r}, t) = \boldsymbol{f}^{ext}, \tag{2.9}$$

$$\nabla \cdot \boldsymbol{u}(\boldsymbol{r}, t) = 0. \tag{2.10}$$

2.2.1 Forces on particles

The equation of motion for particles as used in the present work will be introduced in form of a stochastic differential equation in chapter 3. The section 2.2.1 shall summarize the different forces that may act on a particle. This work deals with purely hydrodynamically interacting non-Brownian particles. The hydrodynamic force acting on particles, especially hydrodynamic interaction, (that means particles interacting via the suspending fluid, cf. (Mewis & Wagner 2013)) plays a significant role. Effects of hydrodynamic interactions, i.e. clustering and lubrication are outlined below. The mathematical derivation of hydrodynamic interaction matrices between particles is summarized in subsection 3.2.1 following e.g. (Durlofsky et al. 1987). In the following subsection, fluid forces (like drag), and non-hydrodynamic and hydrodynamic interparticle forces are summarized from literature. The fluid forces defined here are for small particles in low particle Reynolds number flows (Maxey & Riley 1983). The non-hydrodynamic interparticle forces introduced in this section are also referred to as colloidal interaction and apply to colloidal sized particles (Mewis & Wagner 2013). Since non-Brownian particles shall be compared to Brownian particles, Brownian forces will be introduced in subsection 2.3.1. The present work treats neutrally buoyant particles with a particle density equal to the solvent density. Thus, effects resulting from density differences of particles and surrounding solvent, like particle sedimentation due to gravity, or rising effects in case of a smaller particle density than fluid density (buoyancy effect), cf. (Mewis & Wagner 2013), are excluded.

Fluid forces

Fluid forces acting on a single spherical particle are given in detail in (Crowe et al. 2011) following Maxey & Riley (1983). In the following, the equation of motion for a small spherical particle in a non-uniform unsteady low Reynolds number flow is summarized as given in (Maxey & Riley 1983). A very detailed description can also be found in (Marshall & Li 2014). Fluid force contributions onto a particle comprise steady state drag, lift force, virtual mass term (Crowe et al. 2011) (in other sources referred to as added mass, e.g. (Marshall & Li 2014)), a Basset term (or Basset history term), cf. (Basset 1961, Chapter 22), or contributions from the undisturbed flow, e.g. pressure gradient, see (Marshall & Li 2014), (Crowe et al. 2011).
In the following, the above mentioned forces are outlined according to (Maxey & Riley 1983) if not cited otherwise. When there is no acceleration of the relative particle velocity to the surrounding fluid velocity $\boldsymbol{u}^{fluid}$, the drag force onto the particle in a velocity field is steady (Crowe et al. 2011). The result is Stokes drag as:

$$\boldsymbol{F}^H = -6\pi\eta a(\boldsymbol{U}(t) - \boldsymbol{u}^{fluid}(\boldsymbol{x}_0, t)). \tag{2.11}$$

$\boldsymbol{U}(t)$ is the particle velocity. $\boldsymbol{u}^{fluid}(\boldsymbol{x}_0, t)$ is the undisturbed fluid velocity at the particle center $\boldsymbol{x}_0$. The steady state drag may have an additional term in case of non-rectilinear shear flow to account for curvature in the underlying flow field, which is called Faxén force (Maxey & Riley 1983), (Crowe et al. 2011), cf. (Faxén 1922). But this does not apply to the shear flow according to equation 2.1, consequently there is not such a force, cf. (Crowe et al. 2011). Note, that in the case of many hydrodynamically interacting particles there may be Faxén forces due to the disturbance in the flow field due to the other particles (Durlofsky et al. 1987), (Batchelor & Green 1972b), see section 3.2.

Lift forces originate from particle rotation and can be differentiated into Saffman lift force (due to a velocity gradient such as in shear flow) and a Magnus force (caused by an intrinsic rotation of the sphere), see (Crowe et al. 2011), (Marshall & Li 2014), (cf. also (Magnus 1852)). The lift force due to a surrounding flow like shear flow has been described by Saffman (1965), (Saffman 1968). The relation of Magnus lift force to Stokes drag scales as the particle Reynolds number Re_p from equation 2.4, the relation of Saffman lift force to Stokes drag scales as the square root of Re_p and thus, in case of Re_p tending to zero, both are not considered (Marshall & Li 2014).

Added mass and history force only appear in unsteady flows (Crowe et al. 2011). In the equation of motion from Brady & Bossis (1988), introduced in section 3.2, they are not considered. Still, these forces are of importance as will be pointed out in section 3.1.3. The added mass force is an additional force, necessary to accelerate the particle in the fluid, and scales as the density ratio of fluid to particle density ρ_f/ρ_p compared

to the drag force (Marshall & Li 2014) and is given in (Crowe et al. 2011), cf. (Auton, Hunt & Prud'homme 1988):

$$\boldsymbol{F}^{vm} = \frac{\rho_f \frac{4}{3}\pi a^3}{2}\left(\frac{D\boldsymbol{u}^{fluid}}{Dt} - \frac{d\boldsymbol{U}}{dt}\right), \tag{2.12}$$
$$\text{with } \frac{D\boldsymbol{u}^{fluid}}{Dt} = \frac{\partial \boldsymbol{u}^{fluid}}{\partial t} + (\boldsymbol{u}^{fluid}\cdot\nabla)\boldsymbol{u}^{fluid}.$$

The Basset history force scales as the square root of a Reynolds number based on the relative particle velocity to the undisturbed fluid flow ($|\boldsymbol{U} - \boldsymbol{u}^{fluid}|$) when compared to Stokes drag (Marshall & Li 2014). It is given as (Crowe et al. 2011), (Maxey & Riley 1983):

$$\boldsymbol{F}^{bass} = 6a^2\sqrt{\pi\rho_f\eta}\int_0^t \frac{\frac{d}{dt'}(\boldsymbol{u}^{fluid} - \boldsymbol{U})}{\sqrt{t-t'}}dt'. \tag{2.13}$$

This force takes into account the acceleration history and results from an acceleration of the flow relative to the particle (Marshall & Li 2014).

As a summary of this section, for the case of a neutrally buoyant particle with $St \ll 1$ and $Re_p \ll 1$, the contributions from the fluid forces given above resulting from steady state Stokes rectilinear shear flow onto a single particle reduce to the Stokes drag.

Non-hydrodynamic interparticle forces

Definitions and descriptions for non-hydrodynamic particle interactions or collisions are taken from (Nijenhuis et al. 2007), cf. (Israelachvili 2011). They differentiate short and long range effects, and hard and soft sphere interaction. A particle can have short and long range effects on its surrounding whereby this range can be interpreted as a layer around this particle where the effects are active. The difference between hard and soft sphere interaction is that in the case of hard sphere interaction the particles cannot come closer than this range around them whereby in the latter case, they can enter the range around another particle which results in a soft sphere interaction. Interparticle forces are described by an interaction potential $\Phi_{pot}(r)$ whereby the resulting force is derived via the gradient of this potential:

$$F^P(r) = \frac{d\Phi_{pot}(r)}{dr}, \tag{2.14}$$

with r as the distance between the particle centers, see (Mewis & Wagner 2013). See also (Dhont 1996) for more information. The interparticle forces have influence on the stability of the dispersion in the sense that attractive forces accelerate aggregation (see above for aggregates) and repulsive forces retard aggregation, cf. (Mewis & Wagner 2013), (Nijenhuis et al. 2007). Van der Waals forces (Kruyt, Jonker & Overbeek 1952) are mostly attractive forces (Hamaker 1937) in contrast to electrostatic repulsion. The theory of DLVO (Derjaguin & Landau (1941), and Verwey & Overbeek (1999)) combines these

two interaction potentials to a total potential as the sum of these two. Other possible interaction forces are steric forces, electrosteric forces and depletion forces which are not further discussed here. The simplest way of modeling colloidal interactions may be by a hard sphere potential such that a repulsive force is zero until particles touch and then is set to infinity (Mewis & Wagner 2013), (Nijenhuis et al. 2007):

$$\Phi_{rep}(r) = \begin{cases} \infty & r \leq a \\ 0 & r > a. \end{cases}$$

This is also called excluded volume (Nijenhuis et al. 2007). The simplest way to model electrostatic forces is a hard sphere potential whereby the particle radius is replaced by an effective radius where the electrostatic repulsion is active (Mewis & Wagner 2013).

For particle sizes of a few micrometers (non-colloidal particles) such interparticle forces as electrostatic can often be ignored (Mewis & Wagner 2013). Further, a shear flow may dominate over the interaction forces, see (Brady & Bossis 1988) for a dimensionless measure of shear to interparticle forces. For the theoretical considerations of the idealized case in the present work, it is assumed not to have such non-hydrodynamic interparticle forces in the non-Brownian case.

Hydrodynamic interparticle forces

Hydrodynamic interaction between particles means that the particles influence each other via the suspending fluid, cf. (Dhont 1996), (Mewis & Wagner 2013). At long range, this interaction is a many-body interaction, cf. (Brady & Bossis 1988). At close contact there are lubrication forces, cf. (Russel et al. 1991). Short range lubrication mostly is a two-body interaction whereby the force approaches infinity the closer the particles get, which means that the particles never really touch each other (Russel et al. 1991), (Brady & Bossis 1988), (Mewis & Wagner 2013). It results from a thin layer of fluid that will always be between approaching particles whereby the relative velocity of the close particles tends to zero and the particles may stuck together (Brady & Bossis 1988). Particles in very close contact may stay correlated and construct so-called hydroclusters (Wagner & Brady 2009). Such hydroclusters are aggregates of particles whereby Brownian motion destroys the clusters; thus, the building of hydroclusters is a phenomenon occurring at higher Péclet numbers (Bossis & Brady 1989). Further, clusters can be broken by a repulsive force which has been introduced in numerical simulations to prevent purely hydrodynamically interacting non-Brownian particles from aggregating into infinite clusters (Sierou & Brady 2004). Ball & Melrose (1995) report the occurrence of unphysically narrow gaps between the particles in the absence of any surface interactions or Brownian motion. The problem of preventing particle overlaps of close particles has also been investigated e.g. by Ekiel-Jezewska, Gubiec & Szymczak (2008). Sierou & Brady (2004) use a repulsive force introduced in (Bossis & Brady 1984). The repulsive force shall model residual Brownian motion or surface roughness effects, but may have influence on binary collisions of particles (Drazer, Koplik, Khusid & Acrivos 2002). For the theoretical considerations, the ideal case of

purely hydrodynamically interacting particles in shear-induced diffusion is regarded. The formation of infinite hydroclusters or particle overlaps is excluded and thus a repulsive force is not necessary.

2.3 Brownian particles versus non-Brownian particles

The difference between Brownian and non-Brownian particles will be outlined in this section. More important, the difference between Brownian and non-Brownian diffusion is subject of section 2.3.3. In order to specify non-Brownian particles which are central for the present work, Brownian particles are introduced in section 2.3.1. Systems may comprise specific time scales which determine the processes that occur. The three elementary time scales referring to the physical phenomena taking place in the context of this thesis are the following. For the definition of the time scales and the nomenclature, see also (Brady & Bossis 1988), (Sierou & Brady 2004), (Dhont 1996):

$$\tau_p = \frac{m}{6\pi\eta a}, \quad \text{the inertial relaxation time scale,} \tag{2.15}$$

$$\tau_f = \frac{1}{\dot{\gamma}}, \quad \text{the flow time scale, with the shear rate } \dot{\gamma}, \tag{2.16}$$

$$\tau_D = \frac{a^2}{D_0}, \quad \text{the diffusive time scale, with } \; D_0 = \frac{kT}{6\pi\eta a}. \tag{2.17}$$

m is the mass of one particle. The Stokes-Einstein diffusivity D_0, which captures the diffusivity of an isolated Brownian particle, will further be explained in the section 2.3.3. kT is the thermal energy with k as the Boltzmann constant and T as the absolute temperature. These time scales serve to create dimensionless parameters. The influence of particle inertia is determined by the Stokes number, cf., e.g., (Mewis & Wagner 2013):

$$St = \frac{\tau_p}{\tau_f}, \tag{2.18}$$

i.e. the ratio of the inertial relaxation time to the shear time scale. In the literature, various authors investigated the influence of inertia in terms of finite Stokes numbers, e.g. Drossinos & Reeks (2005) and Subramanian & Brady (2006). The present work is limited to τ_p considerably smaller than τ_f, and hence particle inertia shall be neglected. The Stokes number can be referred to the particle Reynolds number (equation 2.4) via:

$$St = \frac{2}{9}\frac{\rho_p}{\rho_f} Re_p. \tag{2.19}$$

Non-Brownian particles do not perform Brownian diffusion. Particles outside the colloidal regime, i.e. non-colloidal particles which may be just a few micrometers are non-Brownian (Mewis & Wagner 2013) but also particles that do not perform Brownian motion because e.g. a shear flow dominates over the Brownian motion, cf. (Foss & Brady 1999). The size range from colloid to non-colloid is smooth (cf. section 2.1.1) so the description to classify non-Brownian particles in the present work is via the Péclet

number. The ratio of τ_D to τ_f defines the Péclet number, cf. (Mewis & Wagner 2013, p. 81):

$$Pe = \frac{\tau_D}{\tau_f} = \frac{6\pi\eta a^3\dot{\gamma}}{kT}. \tag{2.20}$$

As the Péclet number may also be interpreted as the ratio of shear forces to Brownian forces, the suspension is Brownian for a vanishing Péclet number while it refers to a non-Brownian suspension for a Péclet number tending to infinity (Brady & Bossis 1988). The Péclet number as defined in equation 2.20 refers to the length scale of the particle radius. A very important aspect described in (Dhont 1996) is that instead of the particle radius a, the numerator in 2.20 can also contain the distance in shear flow gradient direction between two largely separated Brownian particles to describe relative particle velocities. On this length scale, shear flow could dominate over Brownian motion even though the Péclet number on the length scale of the radius (equation 2.20) is small. However, such cases are not regarded in the present work.

2.3.1 Brownian particles

The origin of Brownian motion, which is well explained in (Dhont 1996), will be the topic of the current section. It was in 1827 that the Botanist Robert Brown found an irregular motion of pollen grains in water due to the molecule pushes of the water molecules onto the pollen grain particles (Brown 1828). This observation was substantiated by the works of Einstein (1906b), Langevin (1908) (translated in (Lemons & Gythiel 1997)), and Perrin (1910). This irregular motion is known as Brownian motion which is a diffusive motion. The Brownian motion could be indicated as thermal motion. A detailed overview on historical facts about the discovery of Brownian motion can be found in (Russel et al. 1991).

In the introduction of (van Kampen 2007) to Brownian motion, it is stated that, following Einstein (1956) and von Smoluchowski (1906), between two observations of the position of a Brownian particle, the velocity changed several times which means that the autocorrelation time of the velocity is much smaller than the time between two observations. This conclusion followed from considerations about the relaxation time for velocities and considerations about the influence of molecule pushes. That means, there is a clear separation of velocity relaxation and configuration change time scale (Dhont 1996). In the nomenclature here, the velocity changes on τ_p (cf. equation 2.15), the configuration of many particles or the position of a single Brownian particle changes on τ_D (cf. equation 2.17), with $\tau_p \ll \tau_D$.

Concerning the size, going back to the size range analysis in the previous section 2.1.1, Brownian particles are in the size range of colloids (Dhont 1996).The justification for the size range of Brownian (colloidal) particles is the following: For a Brownian particle being at least ten times the size of a solvent molecule, the Brownian motion can be described macroscopically via the solvent viscosity and temperature, (cf. D_0 in equation 2.17) (Dhont 1996), which requires a minimum size for a Brownian particle of $1nm$. The large size of the particle in comparison to the molecule size shall ensure

that the mass of the particle exceeds the mass of the solvent molecules sufficiently, (Dhont 1996), (Mewis & Wagner 2013). So on the particle time and length scale, the solvent can be treated as a continuum (Mewis & Wagner 2013). According to Dhont (1996) (cf. also (Russel et al. 1991)), the maximum size limit is around $10\mu m$, because then the Brownian motion is too small compared to the dimensions of the particle but the exact size dimensions are not sharp, other sources give deviating size ranges, see size ranges of colloidal particles in section 2.1.1.

It is important to know that the molecule pushes onto a Brownian particle are on an even much smaller time scale τ_{mol} than the two time scales τ_p and τ_D of a Brownian particle, i.e. $\tau_{mol} \ll \tau_p \ll \tau_D$, (Dhont 1996). That means, no matter on which time scale the Brownian particle is regarded, the molecule pushes can be introduced as a rapidly fluctuating force which is infinitely short correlated (δ-correlated) (Dhont 1996).

So, as a result from interaction with the solvent molecules, there are two forces acting on a single Brownian particle (Dhont 1996), the Brownian force due to random molecule pushes and a friction due to systematic molecule pushes. For a large enough particle (sufficiently larger than the solvent molecules, see above) with a not too large velocity, this friction force is linear proportional to its velocity with the friction coefficient fr as proportionality factor. The friction force or hydrodynamic force (Mewis & Wagner 2013) is also referred to as Stokes drag, according to Stokes from 1851 and reads, see equation 2.11:

$$F^H = -frU, \tag{2.21}$$

whereby here the fluid velocity is set to zero, i.e. there is no imposed shear flow with $fr = 6\pi\eta a$ as Stokes friction coefficient (Dhont 1996). The friction determines the velocity relaxation time scale τ_p, i.e. the time for an initial velocity to damp out, see equation 2.15, (Risken 1989). In comparison to that, the time scale τ_D describes the time for a Brownian particle to move a distance comparable to its radius, cf. (Mewis & Wagner 2013). The other force in the balance is the Brownian force which can be described via the thermal energy as (Mewis & Wagner 2013):

$$F^B = \frac{kT}{a}, \tag{2.22}$$

with kT as thermal energy. When the mass of the Brownian particle is too large, this force has no influence, (Risken 1989). Note that equation 2.22 describes the magnitude of the Brownian force on τ_D, this will be shown in more detail in chapter 3.1.3. For the final equation of motion as a stochastic differential equation, refer to section 3.1.3 and the sources cited in there. An equation of motion for a probability density function can be found in section 4.3, see (Risken 1989) and other sources in the next chapter. The equations given above are for a single Brownian particle. In case of hydrodynamically interacting Brownian particles, the friction coefficient is extended to a matrix containing the influence of the particles onto each other which is summarized in section 3.2 following (Durlofsky et al. 1987), (Brady & Bossis 1988). Brownian diffusion will be introduced in section 2.3.3.

2.3.2 Non-Brownian particles in shear flow

In the following, non-Brownian particles in shear flow with a Péclet number (equation 2.20) tending to infinity are regarded. The particles shall interact purely hydrodynamically. The subsequent organization with regard to dilute, semi-dilute and dense follows Mewis & Wagner (2013).

In dilute systems of non-Brownian particles, possible hydrodynamic interaction between particles can be neglected and the particles follow the imposed shear flow (Mewis & Wagner 2013). In Stokes flow, the streamlines around a single particle exhibit a fore-aft symmetry, i.e. symmetry with respect to a plane through the particle center and perpendicular to the flow direction, in close proximity to the particle there are closed streamlines, see e.g. (Mewis & Wagner 2013), (Morris 2009).

In the case of semi-dilute suspensions with ϕ around 0.05 - 0.1 up to 0.1-0.15, two-particle interactions can take place occasionally, whereby the trajectories that particles can take when passing each other are related to the streamlines, mentioned above (Mewis & Wagner 2013). Without Brownian motion or particle roughness, in the case of purely hydrodynamically interacting smooth particles, the particle pair trajectories are symmetric and reversible (Morris 2009), (Brady & Morris 1997). Two-particle interaction has been fundamentally investigated by Batchelor & Green (1972a), Batchelor & Green (1972b). When particles come too close, they exhibit lubrication forces, see section 2.2.1.

The fore-aft symmetry and reversibility can be refracted, respectively destroyed by many effects (Morris 2009), (Acrivos, Batchelor, Hinch, Koch & Mauri 1992), (Sierou & Brady 2004), (Wang, Mauri & Acrivos 1996), including surface roughness (Da Cunha & Hinch 1996), (Rampall, Smart & Leighton 1997), residual Brownian motion and interparticle forces (Brady & Morris 1997), and inertia (Subramanian & Brady 2006). Drazer et al. (2002) suggest that also in the case of purely hydrodynamically interactions, the reversibility can be refracted whereby they use a repulsive force in their numerical simulations. There are discussions whether the pure hydrodynamic limit is singular (Brady & Morris 1997). Here, the present work follows the assumptions from (Sierou & Brady 2004) and (Wang et al. 1996) that enough many-body interactions induce irreversible behavior, even though in numerical simulations a repulsive force is used, see e.g. (Sierou & Brady 2004). Many-body interactions occur in the dense case at high enough particle volume fractions.

The occurring phenomenon is called shear-induced diffusion and has first been observed experimentally by Eckstein et al. (1977), Leighton & Acrivos (1987a), and later Breedveld et al. (2002). It is important to keep in mind that experimental conditions can not generate purely hydrodynamically interacting particles without any other influence factors (Mewis & Wagner 2013). Shear-induced diffusion has been investigated numerically by Bossis & Brady (1987), Drazer et al. (2002), Marchioro & Acrivos (2001), Phung, Brady & Bossis (1996) using the Stokesian dynamics method (Brady & Bossis 1988), see also section 3.2; and by Sierou & Brady (2004) using accelerated

Stokesian Dynamics method (Sierou & Brady 2001). See also (Pine, Gollub, Brady & Leshansky 2005) for more information.

It is important to note that it turned out that this shear-induced diffusive behavior is a long-time diffusion whereby for short times, the behavior is deterministic (Sierou & Brady 2004). In the non-Brownian case, the flow time scale $\tau_f = 1/\dot{\gamma}$ is the only relevant time scale, that means there is no separation of time scales as in the Brownian case (Sierou & Brady 2004), (Subramanian & Brady 2004). Thus, the configuration and the velocity (dependent on configuration) change on the same time scale, see (Sierou & Brady 2004). The intensity of the position diffusion coefficients and the exact time for the diffusive behavior to start are dependent on the particle volume fraction, see (Pine et al. 2005), (Santamaría-Holek et al. 2009b), (Sierou & Brady 2004). The present work treats the case that the particle volume fraction is large enough that shear-induced diffusion arises. For numerical simulations, too high volume fractions close to the maximum packing fraction ϕ_{max} need too short time steps in order to resolve the lubrication (Mewis & Wagner 2013), (Melrose & Ball 1995). This case is not regarded in the present work.

2.3.3 Brownian and non-Brownian diffusion: An introduction

From deterministic behavior, it is expected a quadratically growing mean square displacement (Dhont 1996), (Risken 1989) for $\triangle r = r(t_2) - r(t_1)$:

$$\langle(\triangle r)^2\rangle = U(t_1)^2(\triangle t)^2, \tag{2.23}$$

under the condition that U does not change during $\triangle t$ and takes the sharp value $U(t_1)$ at t_1. The $\langle\rangle$-brackets denote an average which can be either an average over all particles in the system or an average over several realizations of the system, i.e. an ensemble average. For diffusive motion can be found the following behavior:

$$\langle(\triangle r)^2\rangle = 2D\triangle t, \tag{2.24}$$

$$\text{respectively } D = \lim_{t\to\infty} \frac{1}{2}\frac{d}{dt}\langle r^2\rangle, \tag{2.25}$$

with $r(0) = 0$, see e.g. (Dhont 1996), (Mewis & Wagner 2013), cf. also equation 3.25 in section 3.1.3. D is the diffusivity, also called diffusion coefficient (there also can be defined particular long and short time diffusion coefficients (Brady & Bossis 1988)). The scope of the present work is self-diffusion which stands in contrast to gradient diffusion (Dhont 1996): A single particle under the influence of other particles or molecules performs self-diffusion. A system of many particles diffusing simultaneously due to a gradient (e.g. in concentration) performs gradient diffusion. This is also referred to as collective diffusion. Gradient diffusion occurs due to a concentration gradient (or density gradient) when particles move to regions with lower concentration. Gradient diffusion can be described in an analog form to Fick's law with a gradient diffusion coefficient, see e.g. (Dhont 1996), (Russel et al. 1991). Further in case of a gradient in the shear rate, there has been found a migration from high to low shear rate regions,

(Mewis & Wagner 2013), see (Leighton & Acrivos 1987b), and (Morris 2009) for a review. Both effects are not considered in the present work. At first will be outlined the Brownian self-diffusion. The description of this behavior shows similarities to the shear-induced self-diffusion of non-Brownian particles, which will be introduced afterwards.

2.3.3.1 Brownian particles

In equation 2.17 was already defined the Stokes-Einstein diffusivity of a single Brownian particle. Related to equation 2.24, this yields:

$$\langle(\triangle r)^2\rangle = 2D_0\triangle t. \tag{2.26}$$

The summarized form over the three dimensions x, y, z of this equation yields a prefactor 6 instead of 2, see (Mewis & Wagner 2013), (Russel et al. 1991). For times $\tau_p \ll t < \tau_D$, Brownian particles exhibit a so-called short-time self-diffusivity (Dhont 1996). For times $\tau_D < t$, there is a long-time diffusivity (Dhont 1996), (Foss & Brady 1999), (Morris & Brady 1996). For arbitrary times t, both in the short-time diffusive regime and in the long-time diffusive regime, use of equation (2.26) requires time steps $\tau_p \ll \triangle t < \tau_D$ to capture the short-time diffusivity of position. For time steps $\triangle t < \tau_p$, respectively $t < \tau_p$, the velocity of the Brownian particle is still correlated because an initial velocity is not damped out yet (Risken 1989). In this regime, equation 2.23 applies, the mean square displacements ($\langle(\triangle x)^2\rangle$, $\langle(\triangle y)^2\rangle$, $\langle(\triangle z)^2\rangle$, and $\langle xx\rangle$, $\langle yy\rangle$, $\langle zz\rangle$) grow quadratically in $\triangle t$ respectively in time t, the position is not diffusive (Bakunin 2011), (Dhont 1996), (van Kampen 2007). Instead, on τ_p, the mean square velocity $\langle U_i(t)U_j(t)\rangle$ for $i, j = x, y, z$ grows linearly in time (see (Risken 1989)). For $t \to \infty$ on τ_p, the mean square velocity reaches a constant equilibrium value and does not grow linearly in time anymore (see (van Kampen 2007)). This will be referred to in section 3.1.3 again. The separation of time scales means: On τ_p the velocity is still correlated but the position is not diffusive (quadratically in time growing mean square displacements), whereby on τ_D, the velocity is uncorrelated but the position of the particle changes via diffusion (linearly in time growing mean square displacements). Note that in the case of many Brownian interacting particles, there is a diffusion matrix $\boldsymbol{D}$ instead of a single coefficient D_0. This diffusion matrix includes the hydrodynamic interaction of all particles onto each other via the inverse of a hydrodynamic resistance matrix $\boldsymbol{R}_{FU}$ instead of the inverse friction coefficient, thus,

$$\boldsymbol{D} = kT\boldsymbol{R}_{FU}^{-1}, \tag{2.27}$$

see (Brady & Bossis 1988), see also section 3.2.

For non-interacting Brownian particles, the long- and short-time diffusion coefficients both are D_0 (Dhont 1996). For interacting Brownian particles, at short times there is hydrodynamic interaction with the surrounding particles, at longer times the particle passes by the surrounding particles and exchanges positions with them (Russel et al. 1991), (Brady & Bossis 1988). As the exchange of positions usually slows the particle

down, the long-time diffusivity is usually smaller than the short-time diffusivity (Brady & Bossis 1988), (Dhont 1996).

2.3.3.2 Non-Brownian particles

For purely hydrodynamically interacting non-Brownian particles in shear flow, deterministic behavior according to equation 2.23 would be expected. Nevertheless, experiments and simulations as outlined in section 2.3.2 indicate the opposite. Sierou & Brady (2004) show simulation results for non-Brownian particles with shear flow in x-direction. They present a linear behavior in time for times $t \to \infty$, i.e. $t > \tau_f$, for the mean square displacements in y- and z-direction, with initial positions of the particles in the origin (Sierou & Brady 2004):

$$\langle yy \rangle \sim 2D_{yy}t, \tag{2.28}$$

$$\langle zz \rangle \sim 2D_{zz}t. \tag{2.29}$$

Here, the $\langle\rangle$-brackets denote an average over all particles in the system. These equations denote diffusive behavior, cf. equation 2.25. The equations for the diffusion components in x-direction, i.e. D_{xx} and D_{xy} are more complicated because of the shear flow in x-direction ((Sierou & Brady 2004) who refer to (Elrick 1962)):

$$\langle xx \rangle = 2D_{xx}t + 2D_{xy}\dot{\gamma}t^2 + \frac{2}{3}D_{yy}\dot{\gamma}^2t^3, \tag{2.30}$$

$$\langle xy \rangle = 2D_{xy}t + D_{yy}\dot{\gamma}t^2. \tag{2.31}$$

$D_{xy} = D_{yx}$ is the only off-diagonal component here (Brady & Morris 1997), (Sierou & Brady 2004). The dimensionless diffusivity scales as $\boldsymbol{D} = a^2\dot{\gamma}\tilde{\boldsymbol{D}}(\phi)$, see (Breedveld 2000), (Breedveld et al. 2002).
The D_{ij} coefficients ($i, j = x, y, z$) in the non-Brownian context include averages over all particles, they do not present the position of each particle in the system anymore. In this way, they are not comparable to the Brownian N-particle diffusion tensor $\boldsymbol{D}$ in equation 2.27 mentioned above, which includes diffusion components for every Brownian particle in the system.

As mentioned in section 2.3.2, the exact time t for shear-induced diffusion to arise depends on the particle volume fraction and may not be exactly $t = \tau_f$, see e.g. (Sierou & Brady 2004). For short times, $t < \tau_f$, the mean square displacements show quadratic behavior in time according to equation 2.23 as shown by Sierou & Brady (2004). The shear-induced diffusion only appears for $t \to \infty$ on τ_f, see e.g. (Breedveld et al. 2002), (Breedveld, van den Ende, Jongschaap & Mellema 2001b), (Drazer et al. 2002), (Sierou & Brady 2004). In this way, it exhibits a difficulty in comparison to Brownian diffusion, because this corresponds to the long-time diffusivity in the Brownian case. As explained by Sierou & Brady (2004), the beginning of the linear behavior at times $t > \tau_f$ denotes the beginning of the diffusive behavior of position with a constant diffusion coefficient. Diffusive behavior begins after enough particle-particle interactions have

taken place and the velocity is not correlated anymore. The difference to the Brownian case is that here is no separation of time scales. In the Brownian case above, the velocity is uncorrelated on the time scale of configurational changes, i.e. $\tau_p \ll \tau_D$, whereby in the non-Brownian case with only one time scale τ_f, the velocity is only uncorrelated for long times $t > \tau_f$. As will be pointed out later, this poses difficulties in the stochastic treatment of non-Brownian particles. In experiments, Breedveld et al. (2001b) also indicate a different kind of diffusion for the small and intermediate time scales which is considerably smaller than the shear induced diffusion. Thus, the present work follows the simulation results from Sierou & Brady (2004) with a non-diffusive behavior of particle positions for short times.

So at arbitrary times t (including times $t > \tau_f$), time steps of $\triangle t > \tau_f$ capture the diffusive behavior in equation 2.24 while time steps $\triangle t < \tau_f$ represent behavior according to equation 2.23.

2.4 Effective viscosity

The behavior of particle flows is strongly influenced by the interaction between the particles which also may influence the rheology of the particle flow. A Newtonian solvent may result in a non-Newtonian suspension due to the particles (Nijenhuis et al. 2007). In case of particles suspended in the solvent, in addition to the solvent viscosity η, there also is an effective viscosity due to the influence of the particles, see e.g. (Mewis & Wagner 2013) for an overview. Further, the particles and the solvent shall have equal density, cf. section 2.2.1. As noted in section 2.1.1, the solvent can be treated as a continuum in the present work. The prerequisite for measuring an effective viscosity is that also the effective fluid, i.e. the solvent containing the particles, can be regarded as a continuum. This is only allowed on a length scale which is large compared to the length scale of the particles (Dhont 1996). According to Stickel & Powell (2005), for particles with a diameter of less than a few micrometers length scales around 100 particle radii suffice to treat the suspension as a continuum with an effective viscosity.

This section is based on (Mewis & Wagner 2013) if not cited otherwise. Mewis & Wagner (2013) give an introduction to the concepts of rheology, cf. also the sources cited in there. For the simple shear flow, in the absence of turbulence (as regarded here), the fluid flow is parallel to the plates, with a gradient of velocity $du_x/dy = \dot{\gamma}$. Newton's constitutive equation relates the stress to shear rate and viscosity. The stress matrix $\boldsymbol{\sigma}$ is contained in the Navier-Stokes equation 2.5 with 2.6 as follows:

$$\nabla \cdot \boldsymbol{\sigma} = -\nabla p + \eta \nabla^2 \boldsymbol{u} \tag{2.32}$$

$$\text{with} \quad \boldsymbol{\sigma} = -p\boldsymbol{I} + \eta[\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T], \tag{2.33}$$

with $\boldsymbol{I}$ as the unity matrix and p as pressure. The superscript T denotes the transpose. For shear flow as regarded here, the only non-zero component of $\nabla \boldsymbol{u}$ is du_x/dy. Thus, $\sigma_{xy} = \sigma_{yx} = \eta\dot{\gamma}$. In case that η is not proportional to the shear rate, Newton's law is not

fulfilled anymore, this yields non-Newtonian behavior. One can distinguish between shear thinning (viscosity decreases with increasing shear rate), shear thickening (viscosity increases with increasing shear rate), and yield stress (non-zero stress for zero shear rate). Time dependent viscosities or memory effects like in viscoelasticity are not considered in the present work.

A relative viscosity is defined as $\eta_r = \eta_{eff}/\eta$. For low Reynolds number flows with monodisperse spherical non-colloidal particles applies $\eta_r = f(\phi)$, for Brownian particles $\eta_r = f(\phi, Pe)$ (Stickel & Powell 2005), (Krieger 1963), (Krieger 1972) which are idealized cases, more influence factors can also be found in (Stickel & Powell 2005), (Krieger 1963), (Krieger 1972).

For dilute non-interacting suspensions, Einstein's formula applies as $\eta_r = (1 + 2.5\phi)$ ((Einstein 1906a), (Einstein 1911), (Happel & Brenner 1965)), which holds irrespectively of particle size, thus for colloids and non-colloids. For low concentrations, the Péclet number dependency of Brownian particles can be neglected. This only applies for Stokes flow, thus small particle Reynolds numbers. For non-negligible Re_p, the particle Reynolds number enters the viscosity equation as $\eta_r = 1 + \phi(2.5 + 1.34Re_p^{1.5})$ as given by (Mewis & Wagner 2013). Following Mewis & Wagner (2013), particle interactions are negligible up to $\phi = 0.05$. The interaction of two or more particles introduces an expansion in the particle volume fraction, where power 2 represents the interaction of particle pairs, and so on: $\eta_r = 1 + 2.5\phi + c_2\phi^2 + c_3\phi^3 \ldots$.
c_2 has been determined for several physical situations, e.g. by Batchelor & Green (1972a). For very high volume fractions, e.g. the maximum packing fraction ϕ_{max} enters the equation. Further information on rheology can be found in (Stickel & Powell 2005) and (Mueller, Llewellin & Mader 2010 (online 2009)).

The derivation of the higher order coefficients poses difficulties for non-Brownian particles because of the closed trajectories that keep particles correlated. The fore-aft symmetry of purely hydrodynamically interacting particles suggests zero normal stress differences, but there is found a loss of fore-aft symmetry and a loss of reversibility, whereby non-zero normal stress differences appear (originating from anisotropy) (Mewis & Wagner 2013), (Morris 2009). In the pure hydrodynamic case of non-Brownian particles in the absence of effects that may destroy clusters, particles will aggregate into infinite clusters. Therefore, very often the limit of Brownian particles with $Pe \to \infty$ is regarded, or additional effects such as roughness are introduced, see section 2.3.2 above. For higher volume fractions, the description of Brownian particles becomes dependent on the Péclet number such as $\eta_r = f(\phi, Pe)$. The following paragraph is based on (Wagner & Brady 2009). For Brownian particles which are in equilibrium increasing the shear rate, i.e. increasing the Péclet number, is followed by a shear thinning region at around $Pe \approx 1$, and a shear thickening region for high Péclet numbers. The shear thinning results from particles getting organized in the flow. The shear thickening region is the regime of dominant hydrodynamic interaction where particles start to cluster. For an analysis of Brownian particles with increasing Péclet number refer to (Foss & Brady 2000). A very detailed review on this behavior is given in (Morris 2009).

The effective viscosity for higher concentrations including hydrodynamic interactions for an arbitrary number of spheres can be included in the derivation of the hydrodynamic interactions, see (Durlofsky et al. 1987) and (Brady & Bossis 1988).

2.5 Short summary of chapter 2

The present work focuses on non-Brownian spherical solid particles in shear flow, which comprises size ranges of colloids (where a background flow dominates over the Brownian motion) and non-colloids. Further, Stokes flow is considered. Even though non-Brownian particles do not perform Brownian motion, they still exhibit diffusive behavior in shear flow due to hydrodynamic interactions. As pointed out in the literature, a main difference between Brownian and non-Brownian particles is a separation of time scales in the Brownian case and a non-separation of time scales in the non-Brownian case. This chapter closes with a short note on the effective suspension viscosity as it is known from the literature that shear-induced diffusion has influence on the rheological behavior. The following chapter 3 proceeds with the equation of motion for particles.

3 The Langevin equation

In chapter 3, the equation of motion for Brownian and non-Brownian particles, the so-called Langevin equation, is introduced. The Langevin equation for a single Brownian particle is presented mainly based on (Risken 1989). Generally, for the present analysis, a physical regime is regarded where particles are considered to have a larger size and mass than the solvent particles or molecules so that the time scale of molecular collisions τ_{mol} onto the particles is smaller than any other time scale of interest, cf. section 2.3. Further, the equation of motion for many interacting particles is shown following (Brady & Bossis 1988) and (Dhont 1996).

3.1 The Langevin equation: General introduction

Origin is Newton's equation of motion as:

$$m\frac{d\boldsymbol{U}}{dt} = \sum \boldsymbol{F}. \tag{3.1}$$

The Langevin equation, which originally was supposed to describe Brownian particles, is derived from equation 3.1 (Zwanzig 2001). It turned out later, that the so-called "Brownian particle" can be taken as representative of many other situations which can be described by the same mathematics, i.e. some collective property of a macroscopic system (Zwanzig 2001). The Langevin equation is originated by Langevin (1908) (translated in (Lemons & Gythiel 1997)). Essentially, the Langevin equation is Newton's equation of motion for a macroscopic particle where the surrounding molecules (or respectively their influence onto the regarded Brownian particle) are modeled as stochastic force (Dhont 1996).

3.1.1 Microscopic and macroscopic treatment

To classify the regarded processes and to better understand the meaning of modeling fluctuations, Risken's definition about microscopic and macroscopic treatment (Risken 1989) will be shortly summarized. The macroscopic treatment is divided into stochastic and deterministic treatment. Further, Risken's definition of heuristic and rigorous derivation is carefully described (Risken 1989). Note that the following shall only give a sense of the different approaches which is sufficient within the scope of the present thesis.

The microscopic treatment of the equation of motion 3.1 requires a system of equations for all variables. This implies, that in addition to the regarded particle, also the

positions of all surrounding fluid particles, i.e. the molecules in the solvent, would have to be known as functions of time (Zwanzig 2001). It is clearly not advantageous to work with such a system.

The macroscopic description which includes both, the stochastic and the deterministic treatment, can be derived either rigorously or heuristically. The rigorous derivation starts with the microscopic equations. From here, the drift and the diffusion terms should be derived which yields the stochastic description. The deterministic description is gained by just neglecting the fluctuations in the stochastic description. In the deterministic description, the variable of interest at a time t is completely determined by its initial value at time t_0 (Risken 1989). According to Risken, the determination of the drift and diffusion terms out of the microscopic equations is not possible in general. The second way is the heuristic derivation which will be referred to later again. In case that the equation for the deterministic behavior of the macroscopic variables is known, the starting point is this deterministic equation and a stochastic force, denoted as $\psi(t)$ below, is added. The problem here is to find the right definition of this stochastic force which is done heuristically. In the case of Brownian particles, this may be the equipartition theorem (see section 3.1.3). Van Kampen (following (Uhlenbeck & Ornstein 1930)) calls this the *Langevin approach* where the Langevin force $\psi(t)$ has to fulfill the following properties (van Kampen 2007):

- $\psi(t)$ is irregular and unpredictable and can be treated as a stochastic process, see section 4.1.
- $\psi(t)$ is given regardless of the macroscopic variable.
- $\psi(t)$ is Gaussian, see section 4.1.3, with the properties given in equations 3.2 and 3.3 below.

Of course, this specification is only a model for real physical fluctuating forces (van Kampen 2007).

In case of a Gaussian fluctuating force, it suffices to give information on the first and second moment of this force, see section 4.1.3, (van Kampen 2007):

$$\langle \psi(t) \rangle = 0, \tag{3.2}$$

$$\langle \psi(t)\psi(t') \rangle = c\delta(t - t'). \tag{3.3}$$

The fluctuating force $\psi(t)$ is given by a Dirac impulse $\delta(t - t')$. A Dirac impulse is defined by $\delta(t - t') = 0$ if $t - t' \neq 0$ and $\int_{-\infty}^{\infty} \delta(t - t')d(t - t') = 1$ (Russel et al. 1991), (Lighthill 1959). In case that the fluctuating force is correlated via a Dirac impulse, it is called a white noise, in contrast to a non-Dirac correlated noise which is called colored noise (Risken 1989). For completeness, a white noise means that the Fourier transform of the correlation function 3.3 is not dependent on frequency in contrast to colored noise where it does depend on frequency (Zwanzig 2001), (Risken 1989). That means, the white noise, modeled by the Dirac impulse, denotes an infinitely short correlated function whereby the colored noise represents a function with a finite correlation time, see e.g. (van Kampen 2007). The constant c in equation 3.3 has to

be adjusted according to statistical mechanics known beforehand or by assumptions how the fluctuations should scale (van Kampen 2007). According to van Kampen (2007), the Langevin approach works well for linear problems, however, quasilinear and nonlinear problems may cause problems, see next section 3.1.2.

3.1.2 Langevin equation

According to Gardiner (2009), the Langevin equation was the first example of a stochastic differential equation. The general form of the Langevin equation may be nonlinear (Gardiner 2009), (van Kampen 2007):

$$\dot{h} = A(h) + C(h)\psi(t), \tag{3.4}$$

with h as the variable of the system and $\psi(t)$ the randomly fluctuating noise term defined in equations 3.2, 3.3. A and C can be any functions specified to the system. Note that according to Gardiner (2009), a non-zero mean of $\psi(t)$ in equation 3.3 could always be embedded in the function A. In the following, an outline about linear, quasilinear and nonlinear equations of motion will be summarized from van Kampen (2007). He states that the Langevin approach (above) is highly successful in case of linear deterministic equations such as:

$$\dot{h} = Ah + \psi(t). \tag{3.5}$$

Then, the first moment $\langle h \rangle$ fulfills the same equation 3.5 as h whereby the Langevin force $\psi(t)$ is omitted (Zwanzig 2001), for the mathematical description of moments refer to section 4.1.1. Equation 3.5 can be related to a linear Fokker-Planck equation (van Kampen 2007). Fokker-Planck equations are introduced in section 4.2. In case of a nonlinear $A(h)$ but still constant C in equation 3.4, the equation would be called quasilinear. There can also be found a corresponding Fokker-Planck equation, which is quasilinear then. Only in the case of a nonlinear function $A(h)$ and a function $C(h)$ in equation 3.4, the system has no well-defined meaning anymore. This yields the so-called Itô-Stratonovich dilemma (see (Stratonovich 1966) , (Itô 1944), and (Risken 1989) for further details). For more information refer also to the sources concerning the Itô-Stratonovich dilemma given in (van Kampen 2007). This is not further discussed here. The noise in the quasilinear function is additive, in the nonlinear function the noise is multiplicative (van Kampen 2007). The following chapters 5 and 6 treat linear Langevin equations.

In case of a Dirac correlation, the noise source $\psi(t)$ is a white noise. Next, the case that the correlation time of $\psi(t)$ is finite but non-zero, i.e. represents a colored noise, will be summarized from (van Kampen 2007). Starting point is the following equation (van Kampen 2007), (van Kampen 1989):

$$\dot{h} = A(h) + C(h)\psi(t), \tag{3.6}$$

$$\text{with } \langle \psi(t) \rangle = 0 \text{ and } \langle \psi(t)\psi(t') \rangle = \kappa(t - t'), \tag{3.7}$$

where $\kappa(t-t')$ is an autocorrelation function that is not a Dirac impulse. Equations such as 3.7 are called Langevin-*like* equations. They include a colored noise instead of white noise. The correlation time is not zero. A possible model for the noise term $\psi(t)$ is a separate Langevin equation as follows:

$$\dot{\psi} = -\frac{1}{\tau_{corr}}\psi + \xi(t), \tag{3.8}$$

with ξ as a white noise. The Langevin equation 3.8 represents a so-called Ornstein-Uhlenbeck process (Uhlenbeck & Ornstein 1930), see section 4.2.3.2. This colored-noise approach will be essential for the derivation of the alternative Fokker-Planck equation in chapter 6.

3.1.3 The Langevin equation (original form for one Brownian particle)

In the following, the Langevin equation for a Brownian particle in a solvent is outlined. This section is mainly based on (Risken 1989), see also (Uhlenbeck & Ornstein 1930). The equations here are presented in one dimension according to Risken. They can be easily transferred to the three-dimensional case as shown also in (Risken 1989) and very detailed in (Dhont 1996).
The exact equation of motion for a Brownian particle in a solvent would require to solve a system of equations for this particle and all the molecules in the solvent which at a number of the order of 10^{23} molecules is impossible. Thus, the heuristic way following the definition of Risken (1989) from section 3.1.1 is used.

In case that the mass of the Brownian particle is much larger than the mass of a solvent molecule, the time scale of the molecule pushes τ_{mol} is much smaller than τ_p and τ_D which is the prerequisite for the Langevin description (Dhont 1996), see also section 2.3.1. The force onto the Brownian particle is a sum of a friction force F^H in equation 2.21 and a randomly fluctuating force F^B in equation 2.22 (Dhont 1996). In case of not too large velocities, F^H is given by Stokes law. The friction also originates from the collisions with the solvent (Dhont 1996). The fluctuating force F^B can only be regarded in terms of ensemble averages, which means an average over several realizations of the system (Risken 1989).

Using the description by Risken (1989), the first part of the Langevin equation is a deterministic differential equation:

$$m\frac{dU}{dt} = -frU, \tag{3.9}$$

where $\frac{m}{fr} = \tau_p$ is the inertial relaxation time, cf. equation 2.15. $U(t)$ is the velocity of the Brownian particle. The deterministic equation shows that a velocity $U(t)$ is determined by its initial velocity $U(t=0) = U_0$ via:

$$U(t) = U_0 \exp\left(-\frac{t}{\tau_p}\right).$$

An initial velocity damps out on the time scale τ_p.

The addition of the force $F^B(t)$ turns equation 3.9 into a stochastic differential equation. Applying van Kampen's definition of Langevin force (see also section 3.1.1) to F^B yields (van Kampen 2007), (Risken 1989): The force $F^B(t)$ is treated as a stochastic process with its properties regardless of U. The autocorrelation function takes into account that the force varies rapidly due to the molecule collisions:

$$\langle F^B(t) \rangle = 0, \tag{3.10}$$

$$\langle F^B(t) F^B(t') \rangle = \Gamma \delta(t - t'), \tag{3.11}$$

with a constant Γ. With equations 3.10 and 3.11, the process is not fully determined yet. So, it is assumed that F^B is a Gaussian process. Gaussian processes are fully determined by the first two moments, see also section 4.1.3. The Dirac impulse conforms to the instantaneous collisions of the molecules. So, equation 3.11 only applies for times larger than the solvent time scale τ_{mol} (Dhont 1996). The Γ is determined via the so-called equipartition theorem which will be shown below.

The resulting Langevin equation reads, see e.g. (Zwanzig 2001), (Dhont 1996):

$$m\frac{dU}{dt} = -frU + F^B(t), \tag{3.12}$$

$$\text{with } \frac{dx}{dt} = U. \tag{3.13}$$

x is the position of the Brownian particle. $fr = 6\pi\eta a$ is Stokes friction. The position and momentum are stochastic variables now which means that no deterministic solution of the equations 3.12 and 3.13 is possible (Dhont 1996).

For the whole equation follows (Risken 1989):

$$U(t) = U_0 \exp\left(-\frac{t}{\tau_p}\right) + \int_0^t \exp\left(-\frac{(t-t')}{\tau_p}\right) \frac{1}{m} F^B(t') dt', \tag{3.14}$$

and for the velocity correlation:

$$\begin{aligned}\langle U(t_1)U(t_2)\rangle &= U_0^2 \exp\left(-\frac{(t_1+t_2)}{\tau_p}\right) \\ &\quad + \int_0^{t_1}\int_0^{t_2} \exp\left(-\frac{(t_1+t_2-t_1'-t_2')}{\tau_p}\right)\frac{\Gamma}{m^2}\delta(t_1'-t_2')dt_1'dt_2' \qquad (3.15)\\ &= U_0^2 \exp\left(-\frac{(t_1+t_2)}{\tau_p}\right) \\ &\quad + \frac{1}{2}\frac{\Gamma}{m^2}\tau_p\left(\exp\left(-\frac{|t_1-t_2|}{\tau_p}\right) - \exp\left(-\frac{(t_1+t_2)}{\tau_p}\right)\right). \qquad (3.16)\end{aligned}$$

For a stationary state, $\langle U(t_1)U(t_2)\rangle$ does not depend on the absolute values t_1, t_2 but only on time differences $|t_1 - t_2|$ (Risken 1989), (van Kampen 2007). The stationary state is reached for $\frac{t_1}{\tau_p} \gg 1$ and $\frac{t_2}{\tau_p} \gg 1$, when initial velocities are completely damped out (Risken 1989):

$$\langle U(t_1)U(t_2)\rangle = \frac{1}{2}\frac{\Gamma}{m^2}\tau_p \exp\left(-\frac{|t_1-t_2|}{\tau_p}\right), \qquad (3.17)$$

which in this context is also called equilibrium state (van Kampen 2007). Γ is derived via the equipartition law of classical statistical mechanics through the following equations (Dhont 1996), (Risken 1989): The average energy in the stationary state of a Brownian particle is:

$$\begin{aligned}\langle E\rangle &= \frac{1}{2}m\langle U(t)^2\rangle \qquad (3.18)\\ &= \frac{1}{2}m\frac{1}{2}\frac{\Gamma}{m^2}\tau_p. \qquad (3.19)\end{aligned}$$

On the other hand applies:

$$\langle E\rangle = \frac{1}{2}kT. \qquad (3.20)$$

Therefore, Γ can be determined as follows:

$$\Gamma = \frac{2kTm}{\tau_p}. \qquad (3.21)$$

With equation 3.21, the equilibrium value for $t \to \infty$ for the Brownian velocity results in, see (van Kampen 2007):

$$\langle U(\infty)^2\rangle = \frac{1}{2}\frac{\Gamma}{m^2}\tau_p = \frac{kT}{m}. \qquad (3.22)$$

Equation 3.22 is a so-called fluctuation-dissipation theorem, as it relates the fluctuation term Γ to the dissipation fr in τ_p (van Kampen 2007). For the three-dimensional case, the equipartition theorem can easily be adapted (Risken 1989). In case of an imposed shear flow, the equipartition theorem has to be changed, the solvent velocity has to be subtracted, see (Dhont 1996).

The mean square displacement can be derived via integration (Dhont 1996), (Risken 1989):

$$\langle (x(t)-x_0)^2 \rangle = \left\langle \int_0^t U(t_1)dt_1 \int_0^t U(t_2)dt_2 \right\rangle = \int_0^t \int_0^t \langle U(t_1)U(t_2)\rangle dt_1 dt_2 \tag{3.23}$$

$$= \left(U_0^2 - \frac{1}{2}\frac{\Gamma}{m^2}\tau_p \right) \tau_p^2 \left(1 - \exp\left(-\frac{t}{\tau_p} \right) \right)^2 + \frac{\Gamma}{m^2}\tau_p^2 t - \frac{\Gamma}{m^2}\tau_p^3 \left(1 - \exp\left(-\frac{t}{\tau_p} \right) \right). \tag{3.24}$$

The order of averaging and integration have been exchanged in equation 3.23, which according to (Haken 1977) is allowed since the averaging has no influence on the integration. Further, the integration procedure from equation 3.23 to 3.24 requires the following rule (Risken 1989):

$$\int_0^t \int_0^t \exp\left(-\frac{|t_1 - t_2|}{\tau_p} \right) dt_1 dt_2 = 2 \int_0^t \left\{ \int_0^{t_1} \exp\left(\frac{-(t_1 - t_2)}{\tau_p} \right) dt_2 \right\} dt_1$$

$$= 2t\tau_p - 2\tau_p^2 \left(1 - \exp\left(-\frac{t}{\tau_p} \right) \right).$$

Equation 3.24 illustrates the separation of time scales for Brownian particles which was referred to in sections 2.3.1 and 2.3.3.1. For $t \to 0$ on τ_p, equation 3.24 recovers the quadratic behavior of the non-diffusive regime, cf. equation 2.23 in section 2.3.3. U_0 can be a sharp initial value for the velocity. In case that the velocity is in equilibrium distribution initially, U_0^2 takes the value kT/m from equation 3.22. If equation 3.24 is regarded on τ_D, i.e. t is on τ_D, it follows $t \gg \tau_p$. Thus, the dominating term in equation 3.24 results in an equation of the form (Risken 1989):

$$\langle (x(t) - x_0)^2 \rangle = 2Dt, \tag{3.25}$$

which is of the form shown in equation 2.24 in section 2.3.3 and denotes the diffusive regime. From equation 3.21, it follows that the diffusion coefficient is:

$$D = \frac{kT}{m}\tau_p = \frac{kT}{fr}, \tag{3.26}$$

as was already introduced in section 2.3 in the definition of the time scale τ_D, i.e. equations 2.17 and 2.26. Equations 3.25 and 3.26 are the fluctuation-dissipation theorem given by Einstein (1905), (Einstein 1906b), these equations relate the mean square of the fluctuations to the damping fr.

Additional effects may be an external imposed shear flow, which is widely discussed for the case of many interacting particles in section 3.2 based on (Brady & Bossis 1988). Besides an imposed shear flow, there also could be an external force field depending on the current position (Risken 1989):

$$m\frac{dU}{dt} = -frU + F(x) + F^B(t), \tag{3.27}$$

depending on the properties of the force field $F(x)$, this problem requires special treatment as will be pointed out in the context of deriving Fokker-Planck equations corresponding to a Langevin equation in section 4.3.2.

3.1.3.1 Criticism on the use of Stokes friction in the Langevin equation

Several works criticize the use of the Langevin equation as given in equation 3.12, e.g. (Hinch 1975), which will be summarized from literature below. Simulation results e.g. by molecular dynamics in liquid by Rahman (1964), (Rahman 1966) and Alder & Wainwright (1970) revealed a discrepancy in the velocity autocorrelation function as shown in equation 3.17. The simulation results of Alder and Wainwright showed a possible long time behavior of the velocity autocorrelation function as $(t^{-3/2})$ instead of the exponential decay. This is also known as long-time tail (Dhont 1996), (Russel et al. 1991), (Russel 1981).

The explanation lies in the derivation of Stokes drag, whereby the following considerations are taken from (Russel et al. 1991) and (Dhont 1996): The Langevin equation is the equation of motion for the particle on τ_p, i.e. describes the change of velocity on τ_p. The velocity of a Brownian particle relaxes on τ_p. Close to the particle, the fluid flow velocity scales as the particle velocity $\boldsymbol{U}$. It decreases to a smaller value over a distance comparable to the particle radius a. In other words, the vorticity of the fluid flow diffuses on the length scale a. The time scale on which the vorticity diffuses is:

$$\tau_{fluid} = \frac{\rho_f a^2}{\eta}, \tag{3.28}$$

cf. the equation for the Reynolds number 2.4. The fluid flow time scale can be related to the particle relaxation time scale via:

$$\frac{\tau_{fluid}}{\tau_p} = \frac{9}{2}\frac{\rho_f}{\rho_p}, \tag{3.29}$$

which is the same relation as given in equation 2.19 with shear flow. For similar densities of particle and solvent, the fluid time scale τ_{fluid} is comparable to τ_p. Due to that, the Stokes drag fr in the Langevin equation on τ_p is a simplified relation. The value $6\pi\eta a$ for the Stokes drag results from the Stokes equations (creeping flow conditions), see e.g. (Dhont 1996). In this sense, Zwanzig & Bixon (1970) state that it is not consistent to derive a friction coefficient from the equation of steady motion (creeping flow conditions) to describe a change in particle velocity (unsteady motion). The Stokes equation may be used instead of the Navier-Stokes equation, when the convection term is negligible and the time derivative can be neglected. Non-dimensionalizing the Navier-Stokes equation 2.5 with the determining component of the particle velocity U, the time scale $\tau_p = m/fr$ and the particle radius a, reads (Dhont 1996):

$$\frac{\rho_f a^2}{\eta}\frac{fr}{m}\frac{\partial \tilde{\boldsymbol{u}}}{\partial \tilde{t}} + Re_U \tilde{\boldsymbol{u}} \cdot \tilde{\nabla}\tilde{\boldsymbol{u}} = \tilde{\nabla}^2\tilde{\boldsymbol{u}} - \tilde{\nabla}\tilde{p} + \tilde{\boldsymbol{f}}^{ext}. \tag{3.30}$$

As can be shown, the convective term can always be neglected in case of small $Re_U = \rho_f a U/\eta$ (the Reynolds number related to U is used, because there is no shear flow here). The time derivative could only be neglected when the time scale of the fluid $\rho_f a^2/\eta$ would be smaller than the time scale of inertial relaxation for the particle τ_p (Russel et al. 1991). Due to equation 3.29, the time derivative can only be neglected when the particle is much denser than the fluid (Dhont 1996).

For similar densities of particle and solvent, the time derivative may only be neglected on τ_D as can be shown by dimensional analysis (Dhont 1996). That means on τ_p, the unsteady Stokes equation has to be solved which would not result in the well known Stokes drag but a time dependent drag (Russel 1981) (Dhont 1996). Additional effects occurring are added mass and history terms which, according to (Landau & Lifshitz 1963, §24) applied onto the particle (Russel et al. 1991), yields:

$$\boldsymbol{F} = -6\pi\eta a\boldsymbol{U} - \frac{2}{3}\pi\rho_f a^3\frac{d\boldsymbol{U}}{dt} - 6\pi a^2\sqrt{\rho_f\eta/\pi}\int_{-\infty}^{t}\frac{d\boldsymbol{U}}{dt'}\frac{1}{\sqrt{t-t'}}dt', \tag{3.31}$$

cf. also the equations for added mass 2.12 and the basset history term 2.13 with $\boldsymbol{u}^{fluid} = 0$ in section 2.2.1. Taking this into account yields the $t^{-3/2}$ behavior (Russel et al. 1991), cf. also (Hauge & Martin-Löf 1973). Widom (1971) has also calculated the velocity autocorrelation function based on the equation of Landau and Lifshitz, thus incorporating added mass and history terms, and also obtained the $t^{-3/2}$ behavior, for times longer than τ_p, (cf. simulation results by Alder & Wainwright (1970) mentioned above). The velocity autocorrelation function obtained to describe the long-time tails is very extensive as shown in e.g. (Clerx & Schram 1992), (Hinch 1975). Clerx & Schram (1992) have also considered the time dependent linearized incompressible Navier-Stokes equation to incorporate the backflow effects of the fluid. The added mass term in equation 3.31 caused a problem in consistency with the equipartition theorem (Clerx & Schram 1992), that could be solved by Zwanzig & Bixon (1975) by taking into account compressibility effects. A very interesting aspect is that Rahman's results also show a region of negative correlated velocity which could be modeled e.g. by Zwanzig & Bixon (1970) and Berne, Boon & Rice (1966). This effect will be referred to in section 8.2.1 again. It is important to state that the short time Langevin equation as originally defined still gives the right diffusivity (Hinch 1975).
In the next section 3.2, the Langevin equation for many interacting particles is summarized from Brady & Bossis (1988). In this context, Bossis & Brady (1987) state that their derivation does not produce such long-time tail behavior as mentioned above for single Brownian particles. They outline that for the behavior on the time scale τ_D, they describe the motion with use of steady hydrodynamic interaction.

3.2 Langevin equation for many hydrodynamically interacting particles

In the preceding section, the Langevin equation for a single Brownian particle was introduced. In case of many particles in the system, the hydrodynamic interaction

between the particles has to be taken into account. This has been done e.g. by Ermak & McCammon (1978) after Deutch & Oppenheim (1971). In this section, the approach used in the Stokesian dynamics method (Brady & Bossis 1988) will be outlined which can be regarded for low and high Péclet numbers, thus for Brownian and non-Brownian particles.

The following equation will also be the starting point for the theoretical considerations in the present work. From now on, the more-dimensional case will be regarded. The equation of motion for N rigid particles is given according to (Brady & Bossis 1988):

$$\boldsymbol{m}\frac{d\boldsymbol{U}}{dt} = \boldsymbol{F}^H + \boldsymbol{F}^P + \boldsymbol{F}^B, \tag{3.32}$$

$$\text{with } \frac{d\boldsymbol{x}}{dt} = \boldsymbol{U}. \tag{3.33}$$

The system in equation 3.32 consists of $6N$ equations, i.e. 3 degrees of freedom, respectively, for both translational and rotational motion for all N particles in the system. $\boldsymbol{m}$ is the generalized mass and moment-of-inertia matrix. Like in the previous section, $\boldsymbol{U}$ is the particle translational and rotational velocity. $\boldsymbol{F}^H$ is the hydrodynamic force and torque, $\boldsymbol{F}^P$ is an interparticle or external force and torque, and $\boldsymbol{F}^B$ is the Brownian force and torque.

In the present work, only the translational components of equation 3.32 will be regarded and, hence, the rotational degrees of freedom are not considered. This work deals with spherically symmetric particles, that means the orientation is not important, whereby disturbances of surrounding particles due to rotation are still included (Dhont 1996). The limitation to the translational degrees of freedom does not pose a problem (Dhont 1996).
The translational components of the terms in equation 3.32 are not renamed to point out the limitation to the translational regime. In the following, the hydrodynamic force $\boldsymbol{F}^H$, the interparticle or external force $\boldsymbol{F}^P$, and the Brownian force $\boldsymbol{F}^B$ will be introduced. Note that the $\boldsymbol{m}$ reduces to a simple m, since in the translational components applies $\boldsymbol{m} = m \cdot \boldsymbol{I}$, with $\boldsymbol{I}$ as a unity matrix of size $3N \times 3N$. m is the mass of a particle.

The theoretical considerations about the hydrodynamic force $\boldsymbol{F}^H$ following below are taken from (Dhont 1996). The following considerations are for the Brownian particle in general, in the absence of shear flow, but will be transferred to incorporate shear flow below. A Brownian particle induces a disturbance that propagates through the solvent to other particles. As the particles are considered to be large compared to the solvent molecules, cf. section 2.3.1, the interactions can be described by the Navier-Stokes equation or Stokes equation. On time scales much larger than τ_p, i.e. $\tau_D \gg \tau_p$, $\boldsymbol{F}^H$ on the i-th particle is influenced by the instantaneous values of the momentum and position of all particles, thus:

$$\boldsymbol{F}_i^H = \boldsymbol{F}_i^H(\boldsymbol{x}_1(t), \cdots, \boldsymbol{x}_N(t), \boldsymbol{U}_1(t), \cdots, \boldsymbol{U}_N(t)). \tag{3.34}$$

The disturbances are transferred as fast as instantaneously. Due to Dhont, on τ_p it can not be ensured that the hydrodynamic interaction is instantaneous, whereas on

τ_D this assumption is valid. For $Re_U \ll 1$ (cf. equation 2.3), which is one of the key assumptions regarded in the present work, on τ_D, $\boldsymbol{F}^H$ can be reconstructed as follows:

$$\boldsymbol{F}^H = -\boldsymbol{R}_{FU}\boldsymbol{U}, \tag{3.35}$$

as a result of Stokes equation. Note that here the full force $\boldsymbol{F}^H$ is given and not just the components on the i-th particle. The friction matrix $\boldsymbol{R}_{FU}$, denoted as resistance matrix in the following, is determined by the instantaneous positions. On τ_p however, the $\boldsymbol{R}_{FU}$ would be time dependent because the time derivative in the Navier-Stokes equation would need to be taken into account, see equation 3.30 in section 3.1.3.1. Note, that the diagonal components of $\boldsymbol{R}_{FU}$ include both, the friction fr of an isolated particle and added the hydrodynamic influence of the other particles as Dhont (1996) points out. It does not pose any problems to limit the considerations to the translational regime because in case of spherically symmetric particles, the translational motion and rotational motion are not coupled which means that if $\boldsymbol{R}_{FU}$ was built for all 6 degrees of freedom, the submatrices connecting the rotational and translational components would be zero (Dhont 1996).

For N particles in shear flow with $Re_p \ll 1$, the hydrodynamic force can be built up as shown in (Brady & Bossis 1988), (Bossis & Brady 1984), (Brenner & O'Neill 1972), (Kim & Mifflin 1985), cf. (Dhont 1996):

$$\boldsymbol{F}^H = -\boldsymbol{R}_{FU}(\boldsymbol{U} - \boldsymbol{U}^\infty) + \boldsymbol{R}_{FE} : \boldsymbol{E}^\infty. \tag{3.36}$$

The operator ":" is a double contraction which is a summation over two indices. The double contraction applied to the rank 3 tensor $\boldsymbol{R}_{FE}$ and the rank 2 tensor $\boldsymbol{E}^\infty$ yields a vector (cf. (Dhont 1996)). Equation 3.36 is given with 6 degrees of freedom in the cited sources but, as pointed out above, here, only the translational degrees are regarded. $\boldsymbol{U}^\infty$ is the bulk shear flow. In contrast to the shear flow velocity given in equation 2.1, here, $\boldsymbol{U}^\infty$ contains the components for N particles. The translational components of the velocity vector of the bulk shear flow for one particle α are $(\dot{\gamma} y_\alpha, 0, 0)$, with y_α as the y-position of this particle α. $\boldsymbol{E}^\infty$ is the symmetric part of the velocity gradient tensor, or the rate of strain tensor:

$$\boldsymbol{E}^\infty = \frac{1}{2}\begin{bmatrix} 0 & \dot{\gamma} & 0 \\ \dot{\gamma} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \tag{3.37}$$

$\boldsymbol{R}_{FU}$ and $\boldsymbol{R}_{FE}$ are the configuration dependent resistance matrices describing the connection between the force/torque ($\boldsymbol{F}$) and the relative velocity ($\boldsymbol{U} - \boldsymbol{U}^\infty$), as well as ($\boldsymbol{F}$) and the rate of strain ($\boldsymbol{E}^\infty$), respectively. Also here, only the translational components are treated. $\boldsymbol{R}_{FU}$ is used to describe the hydrodynamic interaction of the particles on each other while $\boldsymbol{R}_{FE}$ gives the shear-induced disturbances in the flow field (Dhont 1996). The gradient in $\boldsymbol{U}^\infty$, i.e. $\dot{\gamma}$, introduces a rotation of the particles which influences the surrounding particles. Even though the considerations here are limited to the translational degrees of freedom, the influence of shear-induced

rotation onto the translational motion is included due to $(\boldsymbol{R}_{FE} : \boldsymbol{E}^{\infty})_{\alpha}$, the component of $\boldsymbol{R}_{FE} : \boldsymbol{E}^{\infty}$ acting on the particle α, see (Dhont 1996).

$\boldsymbol{R}_{FU}$ and $\boldsymbol{R}_{FE}$ can be built up such that they contain hydrodynamic far-field many-body interactions and near-field lubrication interactions, see (Brady & Bossis 1988), (Durlofsky et al. 1987) for the full derivation. For non-interacting Brownian particles, in the dilute case, the translational components of equation 3.36 result in (Dhont 1996):

$$\boldsymbol{F}^{H} = -fr(\boldsymbol{U} - \boldsymbol{U}^{\infty}). \tag{3.38}$$

In a physical experiment, effects like particle roughness, residual Brownian motion or DLVO type interactions (cf. section 2.2.1) are present and, hence, it is also mandatory for numerical simulations to introduce an interparticle force $\boldsymbol{F}^{P}$ to model such effects (Bossis & Brady 1984), (Drazer et al. 2002), (Brady & Bossis 1988). In the absence of Brownian motion, the necessity of preventing particles from coming too close or building large clusters gives reason for such an interparticle force in numerical simulations, see e.g. (Sierou & Brady 2004), cf. the paragraph on hydrodynamic interaction in section 2.2.1. The translational component of $\boldsymbol{F}^{P}$ can be given in terms of the total potential energy of all Brownian particles, cf. section 2.2.1 equation 2.14. In (Dhont 1996), the translational component of $\boldsymbol{F}^{P}$ is directly $\nabla_{x_i}\boldsymbol{\Phi}_{pot}(\boldsymbol{x}_1, \ldots, \boldsymbol{x}_N)$ with $\boldsymbol{x}_i$ as the position of the i-th particle and ∇_{x_i} as the gradient with respect to all N particles (with $i = 1, \ldots, N$), whereby this $\boldsymbol{\Phi}_{pot}$ also accounts for external potentials. In the theoretical considerations of the present work, purely hydrodynamically interacting particles are considered and, thus, particles without interparticle forces, so that subsequently $\boldsymbol{F}^{P}$ is set to zero.

The molecule impacts of the surrounding fluid onto a particle result in a randomly fluctuating force, i.e. the Brownian force $\boldsymbol{F}^{B}$ (cf. (Ermak & McCammon 1978)). The Brownian force $\boldsymbol{F}^{B}$ in equation 3.32 is modeled such that it has zero average, i.e. $\langle \boldsymbol{F}^{B} \rangle = 0$, and an infinitely short autocorrelation time (Brady & Bossis 1988):

$$\langle \boldsymbol{F}^{B}(0)\boldsymbol{F}^{B}(t) \rangle = 2kT\boldsymbol{R}_{FU}\delta(t). \tag{3.39}$$

This approach follows the Langevin approach from van Kampen (2007) mentioned in section 3.1.1. The $\langle\rangle$ -brackets are the ensemble average, kT is the thermal energy. $\delta(t)$ is the Dirac correlated white noise which models the infinitely short autocorrelated molecule pushes onto the particle. This is the N-particle correspondent to the 1-particle correlation function shown in equation 3.11. For better understanding, the next subsection 3.2.1 shortly outlines the definitions for the resistance matrices $\boldsymbol{R}_{FU}$ and $\boldsymbol{R}_{FE}$. Equation 3.32 is the starting point in chapter 5 for the new multiple time scale analysis.

3.2.1 Derivation of the hydrodynamic resistance tensors

In equation 3.36, the resistance matrices $\boldsymbol{R}_{FU}$ and $\boldsymbol{R}_{FE}$ were introduced. Brady & Bossis (1988) and Durlofsky et al. (1987) have shown how these matrices can be

achieved incorporating both, many-body hydrodynamic interactions and near-field lubrication effects. This will shortly be summarized here. According to (Bossis & Brady 1987), previous Brownian dynamics simulation works did not regard lubrication and many-body interactions.

Lubrication is a two-body interaction and can be introduced in the resistance formulation in a pairwise additive way. In contrast, the many-body interactions have to be implemented in another type of formulation, namely the mobility formulation. The derivation is restricted to vanishingly small particle Reynolds numbers and is based on Stokes equation (Durlofsky et al. 1987). The resistance formulation is the following:

$$\begin{pmatrix} \boldsymbol{F} \\ \boldsymbol{S} \end{pmatrix} = -\boldsymbol{\mathcal{R}} \cdot \begin{pmatrix} \boldsymbol{U} - \boldsymbol{U}^{\infty} \\ -\boldsymbol{E}^{\infty} \end{pmatrix}, \tag{3.40}$$

with

$$\boldsymbol{\mathcal{R}} = \begin{pmatrix} \boldsymbol{R}_{FU} & \boldsymbol{R}_{FE} \\ \boldsymbol{R}_{SU} & \boldsymbol{R}_{SE} \end{pmatrix}. \tag{3.41}$$

$\boldsymbol{F}$ is the force and torque, $\boldsymbol{S}$ is the stresslet onto the particles. $\boldsymbol{\mathcal{R}}$ is called grand resistance matrix whereby its submatrices relate the force and torque and the stresslet to velocity and the rate of strain, respectively. Transforming equation 3.41 yields a so-called grand mobility matrix $\boldsymbol{\mathcal{M}}^{\infty}$:

$$\begin{pmatrix} \boldsymbol{U} - \boldsymbol{U}^{\infty} \\ -\boldsymbol{E}^{\infty} \end{pmatrix} = -\begin{pmatrix} \boldsymbol{M}_{UF} & \boldsymbol{M}_{US} \\ \boldsymbol{M}_{EF} & \boldsymbol{M}_{ES} \end{pmatrix} \cdot \begin{pmatrix} \boldsymbol{F} \\ \boldsymbol{S} \end{pmatrix}, \tag{3.42}$$

with

$$\boldsymbol{\mathcal{M}}^{\infty} = \begin{pmatrix} \boldsymbol{M}_{UF} & \boldsymbol{M}_{US} \\ \boldsymbol{M}_{EF} & \boldsymbol{M}_{ES} \end{pmatrix}. \tag{3.43}$$

The grand mobility matrix is pairwise additive. Inverting the grand mobility matrix $\boldsymbol{\mathcal{M}}^{\infty}$ introduces many-body far field interactions into the grand resistance matrix. Lubrication forces are included in the resistance matrix by adding a matrix $\boldsymbol{\mathcal{R}}_{2B}$ (Bossis & Brady 1987):

$$\boldsymbol{\mathcal{R}} = (\boldsymbol{\mathcal{M}}^{\infty})^{-1} + \boldsymbol{\mathcal{R}}_{2B} - \boldsymbol{\mathcal{R}}_{2B}^{\infty} \tag{3.44}$$

As the two-body lubrication forces have already been counted in the far field parts, the far field parts of those particle interactions ($\boldsymbol{\mathcal{R}}_{2B}^{\infty}$) which introduce lubrication have to be subtracted now.

3.3 Short summary of chapter 3

In the previous chapter 2, possible forces acting on particles were introduced. In the present chapter 3, the equation of motion for particles is presented. In the form of a stochastic differential equation, this is referred to as Langevin equation. A short literature note is given concerning the use of the stationary Stokes equations. The Langevin equation for many hydrodynamically interacting particles requires the derivation of

the hydrodynamic interactions, see (Brady & Bossis 1988). Before proceeding with the multiple time scale analysis in chapter 5, the next chapter 4 gives an introduction to probability theory.

4 Stochastic: The Fokker-Planck equation

The derivation of a new Fokker-Planck equation for non-Brownian particles is an important part of the present work. In this chapter, an introduction to the underlying probability theory and statistical mechanics from literature is given. The Fokker-Planck equation and the necessary stochastic methods are presented. Further, the Fokker-Planck equation as used for Brownian particles is introduced, this comprises the so-called Smoluchowski equation, the Rayleigh equation and the Kramers equation.

4.1 Basic introduction to Probability theory

At first, the difference between probability distribution (also called probability density) and probability distribution function has to be pointed out. This section is based on (van Kampen 2007), if not cited otherwise. A stochastic multidimensional variable is denoted as $\boldsymbol{X}$ (in one dimension X) and can take values $\boldsymbol{x}$ (or in one dimension x). For x as a continuous one-dimensional set of possible values for X, the probability distribution is defined as a function $P(x) \geq 0$ which fulfills:

$$\int P(x)dx = 1. \tag{4.1}$$

The limits of the integration cover the whole range of x, e.g. for x as a velocity, the integration may be performed from $-\infty$ to ∞. The probability for X to take a value between x and $x + dx$ is $P(x)dx$. On the other hand, the probability distribution function $\mathbb{P}(x)$ indicates the total probability that the stochastic variable X has a value $\leq x$:

$$\mathbb{P}(x) = \int_{-\infty}^{x+0} P(x')dx', \tag{4.2}$$

whereby van Kampen (2007) points out that the upper limit $x + 0$ means that a possible delta peak at x is included in the integral. Note, that the present work focuses on the probability distribution P and not the probability distribution function $\mathbb{P}$ from equation 4.2. The probability distribution P completely defines all statistic properties of the stochastic variable X because P can be used to compute any expectation value by integration (Risken 1989), see below.

4.1.1 Averages, characteristic function, cumulants: One dimension

This section is based on (van Kampen 2007) and (Risken 1989). Averages are an important issue of the present work and will be introduced below. The average, also referred to as expectation value of a function $f(X)$, is given by:

$$\langle f(X)\rangle = \int f(x)P(x)dx, \tag{4.3}$$

whereby in general can be defined:

$$\mu_m = \langle X^m\rangle, \tag{4.4}$$

with μ_m as the m-th moment of X and μ_1 as the mean. The characteristic function $C(u)$ is a moment and cumulant generating function of a stochastic variable X:

$$C(u) = \langle \exp(iuX)\rangle \tag{4.5}$$

$$= \int_I \exp(iux)P(x)dx, \tag{4.6}$$

with u as any real number and I as the range of X which is the set or a subset of real numbers. $C(u)$ is the Fourier transform of $P(x)$ in the range I. i is the imaginary unit. Further, $C(0) = 1$ and $|C(u)| \leq 1$. For the definition of $C(u)$ in terms of moments, the exponential function in equation 4.6 is expanded in a Taylor series. Using equations 4.3 and 4.4 with $f(x)$ as the successively higher order functions of x^k ($k = 0, 1, \dots$) yields equation 4.7, whereby cumulants, denoted as κ_m, are defined through equation 4.8, see (Risken 1989), (van Kampen 2007):

$$C(u) = 1 + \sum_{m=1}^{\infty} \frac{(iu)^m}{m!}\mu_m, \tag{4.7}$$

$$= \exp\left(\sum_{m=1}^{\infty} \frac{(iu)^m}{m!}\kappa_m\right). \tag{4.8}$$

The summand 1 in equation 4.7 corresponds to $m = 0$ in the sum. Note that in equation 4.8 there is not a summand 1 in front of the sum (as in equation 4.7) which ensures that the condition $C(0) = 1$ is still fulfilled. Consequently, the characteristic function is known when all moments are known. The inverse Fourier transform of equation 4.6 can be used to build the probability distribution:

$$P(x) = (2\pi)^{-1} \int_{-\infty}^{\infty} C(u) \exp(-iux)du. \tag{4.9}$$

Equation 4.9 only applies for the range I of x from equation 4.6 (van Kampen 2007). It applies without restrictions when x goes from $-\infty$ to ∞ (Risken 1989).

Cumulants are an important point in the present work. The characteristic function as given in equations 4.7 and 4.8 shows the connection between cumulants and moments.

The explicit derivation of the first cumulants by the moments yields the following, see (van Kampen 2007), (Gardiner 2009), (Risken 1989) and more generally (Prohorov & Rozanov 1969, §3.1):

$$\kappa_1 = \mu_1, \tag{4.10}$$
$$\kappa_2 = \mu_2 - \mu_1^2, \tag{4.11}$$
$$\kappa_3 = \mu_3 - 3\mu_2\mu_1 + 2\mu_1^3, \tag{4.12}$$
$$\kappa_4 = \mu_4 - 4\mu_3\mu_1 - 3\mu_2^2 + 12\mu_2\mu_1^2 - 6\mu_1^4. \tag{4.13}$$

κ_4 will be needed in context of the new reduced Fokker-Planck equation in chapter 7. κ_2 is also commonly known as variance, or square of standard deviation.

4.1.2 Multivariate distributions

In the following, multivariate distributions will be introduced because the Fokker-Planck equation derived in the present work in chapter 6 is a multidimensional Fokker-Planck equation. This section is mainly based on (van Kampen 2007).
Instead of X, there is a $\boldsymbol{X}$ with r components and $P_r(x_1, \ldots, x_r)$. It is possible to reduce the number of variables in the probability distribution by integration according to van Kampen (2007) for an $s < r$:

$$P_s(x_1, \ldots, x_s) = \int P_r(x_1, \ldots, x_s, x_{s+1}, \ldots, x_r) dx_{s+1} \ldots dx_r, \tag{4.14}$$

which is then called marginal distribution. This rule will be used in the context of the reduced equation in chapter 7. In case of prescribed values $x_{s+1}, \ldots, x_r$ for the random variables $X_{s+1}, \ldots, X_r$, the conditional probability distribution is computed, rather than the marginal distribution:

$$P_{s|r-s}(x_1, \ldots, x_s | x_{s+1}, \ldots, x_r). \tag{4.15}$$

The |-sign denotes the condition. The relation to $P_r(x_1, \ldots, x_r)$ is, cf. also Bayes' rule:

$$P_r(x_1, \ldots, x_r) = P_{r-s}(x_{s+1}, \ldots, x_r) P_{s|r-s}(x_1, \ldots, x_s | x_{s+1}, \ldots, x_r). \tag{4.16}$$

In analogy to the one-dimensional averages from subsection 4.1.1 above, the following rule applies for the moments here (van Kampen 2007):

$$\langle X_1^{m_1} X_2^{m_2} \cdots X_r^{m_r} \rangle = \int x_1^{m_1} x_2^{m_2} \cdots x_r^{m_r} P_r(x_1, x_2, \ldots, x_r) dx_1 dx_2 \ldots dx_r. \tag{4.17}$$

Also, the characteristic function may be set up for the multivariate case as given in (Risken 1989):

$$C_r(u_1,\ldots,u_r) = \langle \exp(iu_1X_1 + ... + iu_rX_r)\rangle \tag{4.18}$$

$$= \int \ldots \int \exp(i(u_1x_1 + ... + u_rx_r))P_r(x_1,\ldots,x_r)dx_1\ldots dx_r. \tag{4.19}$$

with $C_r(\mathbf{0}) = 1$ and $|C_r(u_1,\ldots,u_r)| \leq 1$ (Gardiner 2009) (cf. (Feller 1971, ch. XV)). After a Taylor expansion, it follows (van Kampen 2007):

$$C_r(u_1,\ldots,u_r) = \sum_0^\infty \frac{(iu_1)^{m_1}\cdots(iu_r)^{m_r}}{m_1!\cdots m_r!}\langle X_1^{m_1}\cdots X_r^{m_r}\rangle \tag{4.20}$$

$$= \exp\left(\sum_0^\infty{}' \frac{(iu_1)^{m_1}\cdots(iu_r)^{m_r}}{m_1!\cdots m_r!}\langle\langle X_1^{m_1}\cdots X_r^{m_r}\rangle\rangle\right). \tag{4.21}$$

The sum here is a sum over all combinations of letting the $m_1,\ldots,m_r$ run from 0 to ∞. Note that there is a prime at the sum $\sum'$ which means that the summand where all $m_1,\ldots,m_r = 0$ is not included because then the condition $C_r(\mathbf{0}) = 1$ would not be fulfilled anymore. The cumulants in equation 4.21 are represented by the $\langle\langle\ \rangle\rangle$-terms and are defined below in equations 4.24a and the following. After taking the inverse Fourier transform of equation 4.19, it follows (Risken 1989):

$$P_r(x_1,\ldots,x_r) = (2\pi)^{-r}\int\ldots\int \exp\left(-i(u_1x_1 + ... + u_rx_r)\right)C_r(u_1,\ldots,u_r)du_1\ldots du_r. \tag{4.22}$$

Also in the mutlivariate case, it is possible to derive the cumulants as combinations of the moments. The second order moments can be represented in a so-called covariance matrix which obeys the following rule (van Kampen 2007):

$$\langle\langle X_iX_j\rangle\rangle = \langle(X_i - \langle X_i\rangle)(X_j - \langle X_j\rangle)\rangle = \langle X_iX_j\rangle - \langle X_i\rangle\langle X_j\rangle. \tag{4.23}$$

The diagonal elements of the covariance matrix $\langle\langle X_iX_j\rangle\rangle$ are called variances, the off-diagonal elements are called covariances. In normalized form, the off-diagonal elements of 4.23 yield the correlation coefficients. Here will shortly be outlined the definition of correlation for $r = 2$ from (van Kampen 2007) because the correlation of variables will be an important argument in later chapters: In case of zero covariance, two variables X_1 and X_2 are uncorrelated. This is not enough for statistical independence. Statistical independence would be fulfilled if all cumulants $\langle\langle X_1^{m_1}X_2^{m_2}\rangle\rangle$ vanish (with $m_1 \neq 0$ and $m_2 \neq 0$).
In addition, also higher order cumulants will be important in the derivation of the reduced form of the colored-noise Fokker-Planck equation in chapter 7. Gardiner (2009) (cf. (van Kampen 1974)) specifies the cumulants up to fourth order:

$$\langle\langle X_i\rangle\rangle = \langle X_i\rangle, \tag{4.24a}$$

$$\langle\langle X_iX_j\rangle\rangle = \langle X_iX_j\rangle - \langle X_i\rangle\langle X_j\rangle, \tag{4.24b}$$

$$\langle\langle X_i X_j X_k\rangle\rangle = \langle X_i X_j X_k\rangle - \langle X_i X_j\rangle\langle X_k\rangle - \langle X_i\rangle\langle X_j X_k\rangle - \langle X_i X_k\rangle\langle X_j\rangle + 2\langle X_i\rangle\langle X_j\rangle\langle X_k\rangle, \tag{4.24c}$$

$$\begin{aligned}\langle\langle X_i X_j X_k X_l\rangle\rangle = {} & \langle X_i X_j X_k X_l\rangle - \langle X_i\rangle\langle X_j X_k X_l\rangle - \langle X_j\rangle\langle X_k X_l X_i\rangle \\ & - \langle X_k\rangle\langle X_l X_i X_j\rangle - \langle X_l\rangle\langle X_i X_j X_k\rangle \\ & - \langle X_i X_j\rangle\langle X_k X_l\rangle - \langle X_i X_k\rangle\langle X_j X_l\rangle - \langle X_i X_l\rangle\langle X_j X_k\rangle \\ & + 2(\langle X_i\rangle\langle X_j\rangle\langle X_k X_l\rangle + \langle X_i\rangle\langle X_k\rangle\langle X_j X_l\rangle + \dots) \\ & - 6\langle X_i\rangle\langle X_j\rangle\langle X_k\rangle\langle X_l\rangle.\end{aligned} \tag{4.24d}$$

4.1.3 Gaussian distribution

The Gaussian distribution is a very important probability distribution throughout the present work. This section is based on (van Kampen 2007) and (Risken 1989). In the Gaussian distribution, all cumulants except for the first and second cumulant are zero. That means, in the cumulant generating function 4.21, cumulants with $m_1 + m_2 + ... + m_r > 2$ vanish. The multivariate Gaussian distribution reads (van Kampen 2007):

$$P_r(\boldsymbol{x}) = (2\pi)^{-r/2}(Det\ \boldsymbol{\Xi})^{-1/2}\exp\left(-\frac{1}{2}(\boldsymbol{x} - \langle\boldsymbol{X}\rangle)^T\boldsymbol{\Xi}^{-1}(\boldsymbol{x} - \langle\boldsymbol{X}\rangle)\right), \tag{4.25}$$

r is the dimension of $\boldsymbol{x}$. The superscript T denotes the transpose. $\boldsymbol{\Xi}$ is assumed to be positive definite with:

$$\langle\langle X_i X_j\rangle\rangle = \Xi_{ij}, \tag{4.26}$$

cf. equation 4.23. When $\boldsymbol{\Xi}$ is positive definite, the inverse, the square root and the inverse square root exist (Risken 1989). All information necessary for the Gaussian distribution are the first and second moments. The sum of mutually independent Gaussian variables yields again a Gaussian distribution (van Kampen 2007). The so-called central limit theorem enables a way to treat sums of stochastic variables as Gaussian under certain conditions (Haken 1977), (van Kampen 2007).

4.1.4 Incorporating time: Stochastic processes

Due to van Kampen (2007), in the context of physics, the name stochastic process implicates time. Incorporating the time t into the description of stochastic variables yields a stochastic process (van Kampen 2007). An example for a stochastic process is the position variable of a Brownian particle. As before, also this section is mainly based on (van Kampen 2007).

A stochastic process may be given as $Y_X(t) = f(X,t)$ whereby the f represents a mapping of the stochastic variable X and time t to a quantity Y. From that follows the probability density for $Y_X(t)$ as follows:

$$P_1(y,t) = \int \delta(y - Y_x(t))P_X(x)dx, \tag{4.27}$$

which gives the probability density for a value y at time t, given the probability density $P_X(x)$ of X. In the integral, the subscript of $Y_X(t)$ changes to a small x. $Y_x(t) = f(x,t)$ is one realization of the process. In this context, the term ensemble average is introduced. The ensemble average is the average over all samples of a process. One supposes to have a random variable realized in an ensemble of systems (Risken 1989) where for each system the stochastic variable has a certain value depending on time, so one takes averages over the ensemble which depend on time with a probability density defined as:

$$P_1(y,t) = \langle \delta(y - Y_x(t)) \rangle, \tag{4.28}$$

with the $\langle\rangle$-brackets here as the so-called ensemble average. The systems are macroscopically identical but pose different microscopic realizations (Dhont 1996). Further applies (van Kampen 2007):

$$P_n(y_1,t_1;y_2,t_2;\ldots;y_n,t_n) = \int \delta(y_1 - Y_x(t_1))\delta(y_2 - Y_x(t_2))\cdots\delta(y_n - Y_x(t_n))P_X(x)dx, \tag{4.29}$$

$$\langle Y(t_1)Y(t_2)\cdots Y(t_n)\rangle = \int y_1 y_2 \cdots y_n P_n(y_1,t_1;y_2,t_2;\ldots;y_n,t_n)dy_1 dy_2 \cdots dy_n. \tag{4.30}$$

The autocorrelation function is defined as follows (van Kampen 2007):

$$\begin{aligned} \langle\langle Y(t_1)Y(t_2)\rangle\rangle &= \langle (Y(t_1) - \langle Y(t_1)\rangle)(Y(t_2) - \langle Y(t_2)\rangle)\rangle \\ &= \langle Y(t_1)Y(t_2)\rangle - \langle Y(t_1)\rangle\langle Y(t_2)\rangle. \end{aligned} \tag{4.31}$$

In case that P_n (equation 4.29) is not influenced by a change of time from t_i to $t_i + T$, the stochastic process is stationary and instead of total times, time differences $|t_i - t_{i+1}|$ are important (Risken 1989). The autocorrelation time then is defined as the time $\tau_{corr} < |t_i - t_{i+1}|$ for that the autocorrelation function becomes zero or negligible. A Gaussian process premises that all P_n obey Gaussian distributions, respectively multivariate Gaussian distributions. Also for stochastic processes, this means that all cumulants > 2 are zero and that the Gaussian process is determined by its first and second moment. Equation 4.25 remains valid for $P(\boldsymbol{x},t)$ whereby for the solution an initial condition is required, see (van Kampen 2007).

4.2 Introduction to the Fokker-Planck equation

Within this section, the origin of the Fokker-Planck equation shall be summarized from literature, this includes an introduction to Markov processes and the so-called Master equation. Further, this section includes an introduction to the Ornstein-Uhlenbeck process.

4.2.1 Markov processes

In the present work, the Markov property plays an important role in the derivation of the new Fokker-Planck equation in chapter 6. This section is based on (van Kampen 2007) and (Risken 1989).
For Markov processes applies, with $t_1 < t_2 < ... < t_n$:

$$P_{1|n-1}(y_n, t_n|y_1, t_1; \ldots; y_{n-1}, t_{n-1}) = P_{1|1}(y_n, t_n|y_{n-1}, t_{n-1}), \tag{4.32}$$

$$P_3(y_1, t_1; y_2, t_2; y_3, t_3) = P_2(y_1, t_1; y_2, t_2) P_{1|2}(y_3, t_3|y_1, t_1; y_2, t_2) \tag{4.33}$$

$$= P_1(y_1, t_1) P_{1|1}(y_3, t_3|y_2, t_2) P_{1|1}(y_2, t_2|y_1, t_1), \tag{4.34}$$

whereby the |-sign is explained in equation 4.15 section 4.1.2 above and describes the conditional probability. The Markov property of a process allows to construct the whole hierarchy of P_n by only knowing $P_1(y_1, t_1)$ and the transition probability $P_{1|1}(y_2, t_2|y_1, t_1)$.
The probability distribution for a change of the random variable depends on the value of the random variable at the last time step but not on earlier time steps (van Kampen 2007). Referring to equation 4.32, this means that the conditional probability density for a y_n at time t_n depends only on information from the last time t_{n-1} but not from earlier times (Risken 1989). Thus, in contrast to a purely random process, a Markov process implies memory of the variable from the last time the variable was measured. That means, the longer this time difference is, the weaker is the memory. On the other hand, the shorter the time difference is, the sharper is the conditional probability. A non-Markovian process can become Markovian again by taking into account more variables. The process for all variables then may be a Markov process again. Integrating the probability density over one or more variables may destroy the Markov property because the remaining joint probability distribution has lost information through the integration (Risken 1989). A Markov process enables to use small time step sizes $\triangle t$ and determine the long time behavior in a so-called Fokker-Planck equation which will be introduced below (van Kampen 2007). Stationary Markov processes can be used to describe equilibrium fluctuations (van Kampen 2007). A very well known process in that context is the Ornstein-Uhlenbeck process (Uhlenbeck & Ornstein 1930) which will be a very important aspect in the derivation of the model for the colored-noise velocity in chapter 6. The Ornstein-Uhlenbeck process is introduced in section 4.2.3.2.

4.2.2 The Master equation

This section is based on (van Kampen 2007). The Fokker-Planck equation can be derived via the so-called Master equation. Starting point is the Chapman-Kolmogorov equation. The Chapman-Kolmogorov equation is derived from equation 4.34 by integrating over y_2 and dividing by $P_1(y_1, t_1)$:

$$P_{1|1}(y_3, t_3|y_1, t_1) = \int P_{1|1}(y_3, t_3|y_2, t_2) P_{1|1}(y_2, t_2|y_1, t_1) dy_2, \tag{4.35}$$

for $t_1 < t_2 < t_3$. Equation 4.35 is also valid for a vector $\boldsymbol{y}$ with r components. A Markov process is defined through P_1 and $P_{1|1}$ (cf. equation 4.34), whereby $P_{1|1}$ fulfills equation 4.35 and P_1 fulfills the following equation, cf. (Risken 1989):

$$P_1(y_2, t_2) = \int P_{1|1}(y_2, t_2|y_1, t_1) P_1(y_1, t_1) dy_1. \tag{4.36}$$

The Master equation is now derived as the differential form of the Chapman-Kolmogorov equation 4.35. To simplify the derivation, van Kampen assumes a homogeneous Markov process, which means that the transition probabilities are stationary and can be written as $W(y|y')$ (no time variable as in $P_{1|1}$). The result is, see (Risken 1989), (van Kampen 2007):

$$\frac{\partial P(y,t)}{\partial t} = \int (W(y|y')P(y',t) - W(y'|y)P(y,t)) dy'. \tag{4.37}$$

With $W(y|y') = W(y'; r)$ and $r = y - y'$, it follows:

$$\frac{\partial P(y,t)}{\partial t} = \int W(y - r; r) P(y - r, t) dr - P(y,t) \int W(y; -r) dr. \tag{4.38}$$

The transition probability is sharply peaked which means that $W(y'; r) \approx 0$ for $|r| > \delta$, $\delta > 0$. Furthermore, the transition probability should vary only slowly in y' which means that the transition probability for a varied y' should result in $W(y' + \triangle y; r) \approx W(y'; r)$ for $|\triangle y| < \delta$. Altogether, that means that there are only small jumps. Further, the probability density shall also be only varying slowly so that an expansion is possible. In the next step, equation 4.38 is expanded in a Taylor series in r. The result is the Kramers-Moyal expansion (Kramers 1940) (Moyal 1949) as given in (van Kampen 2007):

$$\frac{\partial P(y,t)}{\partial t} = \sum_{n=1}^{\infty} \frac{1}{n!} \left(-\frac{\partial}{\partial y} \right)^n \{a_n(y) P(y,t)\}, \tag{4.39}$$

with $a_n = \int_{-\infty}^{\infty} r^n W(y; r) dr$ as the so-called jump moments. A more general result of such a Taylor expansion is given in (Risken 1989) which contains the time dependence in the transition probability. The result is the Kramers-Moyal expansion as given in (Risken 1989):

$$\frac{\partial P(y,t)}{\partial t} = \sum_{n=1}^{\infty} \left(-\frac{\partial}{\partial y} \right)^n \{D^{(n)}(y,t) P(y,t)\}. \tag{4.40}$$

The $D^{(n)}$ is the highest order term in a Taylor expansion of the moments M_n for small δt:

$$M_n(y,t,\delta t)/n! = D^{(n)}(y,t)\delta t + O(\delta t^2), \tag{4.41}$$

with $M_n(y',t,\delta t) = \langle (Y(t+\delta t) - Y(t))^n \rangle|_{Y(t)=y'} = \int (y-y')^n W(y,t+\delta t|y',t)dy$. There is no δt^0 (δt to the power of 0)-term in equation 4.41, so that for $\delta t = 0$, the transition probability is just the δ-function, and the moments are zero, (Risken 1989). For Markov processes, equation 4.40 is of first order in time and the transition probability also fulfills the same equation 4.40 (Risken 1989). For the multivariable case, the Kramers-Moyal expansion looks as follows (Risken 1989):

$$\frac{\partial P(\boldsymbol{y},t)}{\partial t} = \sum_{n=1}^{\infty} \frac{(-\partial)^n}{\partial y_{j_1} \dots \partial y_{j_n}} \{D^{(n)}_{j_1,\dots,j_n}(\boldsymbol{y},t)P(\boldsymbol{y},t)\}, \tag{4.42}$$

with $M^{(n)}_{j_1,j_2,\dots,j_n}(\boldsymbol{y},t,\tau)/n! = D^{(n)}_{j_1,j_2,\dots,j_n}(\boldsymbol{y},t)\delta t + O(\delta t^2)$. $\boldsymbol{y}$ is the set of variables of interest with dimension j. In this context Risken (1989) cites Pawula (1967) by the statement that the expansion either can be terminated after the first term, after the second term, or requires an infinite number of terms. The Kramers-Moyal expansion will also be referred to in section 8.2.2 again.

4.2.3 Fokker-Planck equation for different time scales

In principle, the Fokker-Planck equation ((Fokker 1914), (Planck 1917)) is a parabolic differential equation for the probability density $P(\boldsymbol{y},t)$, see (van Kampen 2007). It is a Master equation as given in the previous section where the Kramers-Moyal expansion is terminated after the second order (van Kampen 2007). In the following, only the simplified version in terms of equation 4.39 is used which means that the transition probability is stationary. This section is based on (van Kampen 2007) and (Haken 1977).

A Fokker-Planck equation is mathematically equivalent to a Langevin equation. That means in analogy to the linear Langevin equation 3.5 in section 3.1.2, can be defined a linear Fokker-Planck equation based on equation 4.39 with $a_1(\boldsymbol{y}) = c\,\boldsymbol{y}$ (c =constant) and constant a_2. The quasilinear Langevin equation in section 3.1.2 is related to a quasilinear Fokker-Planck equation based on equation 4.39 with an $a_1(\boldsymbol{y})$ and a constant a_2. Concerning the nonlinear Langevin equation 3.4, there exists no unique corresponding Fokker-Planck equation.
The Fokker-Planck equation reads:

$$\frac{\partial P(\boldsymbol{y},t)}{\partial t} = -\sum_{i=1}^{r} \frac{\partial}{\partial y_i}\Big(A_i(\boldsymbol{y})P\Big) + \frac{1}{2}\sum_{i,j=1}^{r} \frac{\partial^2}{\partial y_i \partial y_j}\Big(\mathcal{D}_{ij}(\boldsymbol{y})P\Big). \tag{4.43}$$

Equation 4.43 can be derived by the differential quotient $\triangle P/\triangle t$, with $\triangle P(\boldsymbol{y},t) = P(\boldsymbol{y},t+\triangle t) - P(\boldsymbol{y},t)$ in the limit $\triangle t \to 0$. $\boldsymbol{y}$ has dimension r here.
The drift A_i and diffusion terms $\mathcal{D}_{ij}$ of the Fokker-Planck equation can be found from

the corresponding Langevin equation. $\boldsymbol{y}$ is evaluated during the characteristic time step $\triangle t$, for $\triangle t \to 0$ on the regarded time scale:

$$A_i(\boldsymbol{y}) = \frac{\langle \triangle y_i \rangle_{\boldsymbol{y}}}{\triangle t}, \tag{4.44}$$

$$\mathcal{D}_{ij}(\boldsymbol{y}) = \frac{\langle \triangle y_i \triangle y_j \rangle_{\boldsymbol{y}}}{\triangle t}, \tag{4.45}$$

whereas $\triangle y_i$ is the change of the variable of interest during $\triangle t$, i.e. $\triangle y_i = y_i(t + \triangle t) - y_i(t)$. The $\langle\rangle_{\boldsymbol{y}}$ represents a conditional average with a known constant reference value. In the present work an index $\boldsymbol{y}$ denotes the reference to the current $\boldsymbol{y}(t)$. The time step $\triangle t$ on the regarded time scale has to be small which ensures that $\boldsymbol{y}$ does not change much during $\triangle t$. Due to the condition that $\triangle t \to 0$ on the regarded time scale, all terms with O($\triangle t$) in the calculation of equations 4.44 and 4.45 can be neglected. In the following, only linear Fokker-Planck equations are regarded which, following the definition from van Kampen (2007), means that the drift coefficients are linear functions of $\boldsymbol{y}$ and the diffusion terms $\mathcal{D}_{ij}$ are constant. Further, $\boldsymbol{y}$ is supposed to be a Markov variable. (For non-Markovian processes the use of a Fokker-Planck equation requires to incorporate memory functions (Risken 1989).) Under the Markov process assumption, the change of $\boldsymbol{y}$ during $\triangle t$ then serves to find $\triangle P / \triangle t$, or $\partial P / \partial t$ in the limit $\triangle t \to 0$.

4.2.3.1 Multivariate Fokker-Planck equation

The general r-dimensional Fokker-Planck equation in linear form reads (van Kampen 2007):

$$\frac{\partial P(\boldsymbol{y}, t)}{\partial t} = -\sum_{i,j} A_{ij} \frac{\partial}{\partial y_i}(y_j P) + \frac{1}{2} \sum_{ij} B_{ij} \frac{\partial^2 P}{\partial y_i \partial y_j}. \tag{4.46}$$

This equation can be solved with a Gaussian solution. The initial condition shall be $P(\boldsymbol{y}, 0) = \prod_{i=1}^{r} \delta(y_i - y_{i0})$. The solution is given by (van Kampen 2007):

$$P(\boldsymbol{y}, t) = (2\pi)^{-r/2} (Det\ \boldsymbol{\Xi})^{-1/2} \exp(-\frac{1}{2}(\boldsymbol{y} - \langle \boldsymbol{y} \rangle)^T \boldsymbol{\Xi}^{-1} (\boldsymbol{y} - \langle \boldsymbol{y} \rangle)) \text{ with} \tag{4.47}$$

$$\boldsymbol{\Xi} = \int_0^t \exp((t - t')\boldsymbol{A}) \boldsymbol{B} \exp((t - t')\boldsymbol{A}^T) dt' \tag{4.48}$$

$$\text{and } \langle \boldsymbol{y} \rangle = \exp(t\boldsymbol{A}) \boldsymbol{y}(0) \tag{4.49}$$

with the superscript T as the transpose. Further, for $\boldsymbol{\Xi}$ applies:

$$\Xi_{kl} = \langle \langle y_k y_l \rangle \rangle = \langle y_k y_l \rangle - \langle y_k \rangle \langle y_l \rangle. \tag{4.50}$$

4.2.3.2 The Ornstein-Uhlenbeck process

The Ornstein-Uhlenbeck process (Uhlenbeck & Ornstein 1930) (cf. also (Wang & Uhlenbeck 1945)) has originally been derived for the Brownian velocity and is important for the description of equilibrium fluctuations (van Kampen 2007). This section is based on (Risken 1989).
The Ornstein-Uhlenbeck process describes a stationary Gaussian Markov process (van Kampen 2007). The Langevin equation, corresponding to the Ornstein-Uhlenbeck process in three dimensions, is the following, cf. equation 3.8 in one dimension, see also (van Kampen 2007), for $i = 1, \ldots, N$ (in this context, N is a general placeholder, e.g. with $i = x, y, z$):

$$\dot{\psi}_i + \sum_{j=1}^{N} \gamma_{ij}\psi_j = \xi_i(t), \tag{4.51}$$

with $\langle \xi_i(t) \rangle = 0$ and $\langle \xi_i(t)\xi_j(t') \rangle = q_{ij}\delta(t-t')$ with $q_{ij} = q_{ji}$. The homogeneous equations are linear and q_{ij} is independent of ψ_i. For $N = 1$, equation 4.51 is the one-dimensional equation of motion for a Brownian particle shown in equation 3.12 in section 3.1.3. For $\gamma_{ij} = 0$ this is the so-called Wiener process. Given the initial value $\psi_i(0) = \psi_{0i}$, the homogeneous solution (denoted through the superscript *homogen*) reads:

$$\psi_i^{homogen}(t) = \sum_{j=1}^{N} \exp(-\gamma_{ij}t)\psi_{0j}, \tag{4.52}$$

with $\exp(-\boldsymbol{\gamma}t) = \boldsymbol{I} - \boldsymbol{\gamma}t + \frac{1}{2}\boldsymbol{\gamma}^2t^2 \pm \ldots$, cf. Green's function in (Risken 1989). $\boldsymbol{\gamma}$ is the matrix containing the elements γ_{ij} and $\boldsymbol{I}$ is the identity matrix. The inhomogeneous solution (denoted through the superscript *inhomogen*) reads:

$$\psi_i^{inhomogen}(t) = \int_0^{\infty} \sum_{j=1}^{N} \exp(-\gamma_{ij}t')\xi_j(t-t')dt'. \tag{4.53}$$

Thus, $\psi_i(t) = \psi_i^{homogen} + \psi_i^{inhomogen}$. The first moment is given by:

$$\langle \psi_i \rangle = \sum_{j=1}^{N} \exp(-\gamma_{ij}t)x_j, \tag{4.54}$$

which for small times $t > 0$ results in:

$$\langle \psi_i(t) \rangle = x_i - \sum_{j=1}^{N} \gamma_{ij}x_jt + \frac{1}{2}\sum_{j,k=1}^{N} \gamma_{ik}\gamma_{kj}x_jt^2 \pm \ldots \tag{4.55}$$

The second moment reads:

$$\sigma_{ij} = \langle [\psi_i(t) - \langle \psi_i(t) \rangle][\psi_j(t) - \langle \psi_j(t) \rangle] \rangle = \sum_{k,s=1}^{N} \int_0^t \exp(-\gamma_{ik}t') \exp(\gamma_{js}t')dt'q_{ks}, \tag{4.56}$$

which for small times $t > 0$ yields:

$$\sigma_{ij} = q_{ij}t - \frac{1}{2}\sum_{k=1}^{N}(\gamma_{ik}q_{kj} + \gamma_{jk}q_{ik})t^2 \pm \dots \tag{4.57}$$

Higher moments ($n > 2$) vanish for odd n and are proportional to $t^{n/2}$ for even n. This is due to the fact that cumulants of higher order than 2 vanish for a Gaussian distribution, cf. section 4.1.3, see equations 4.10 to 4.13 for the cumulants with $\kappa_3, \kappa_4, ... = 0$. The correlation function for a $t' > 0$ yields:

$$K_{ij}(t', t) = \langle \psi_i(t + t')\psi_j(t)\rangle = \sum_{s=1}^{N} \exp(-\gamma_{is}t')\langle \psi_s(t)\psi_j(t)\rangle = \sum_{s=1}^{N} \exp(-\gamma_{is}t')K_{sj}(0, t), \tag{4.58}$$

and for $t' \leq 0$:

$$K_{ij}(t', t) = \sum_{s=1}^{N} \exp(-\gamma_{js}|t'|)K_{is}(0, t - |t'|).$$

Equation 4.58 can also be seen in equation 3.16 for:

$$K(t', t) = \langle U(t_1 + t')U(t_1)\rangle = \exp\left(-\frac{|t'|}{\tau_p}\right)\langle U(t_1)^2\rangle.$$

A stationary solution of the Langevin equation 4.51 exists if the eigenvalues of $\boldsymbol{\gamma}$ are positive. Then $K_{ij}(t') = K_{ji}(-t')$, further, equation 4.58 does not depend on the total time t and so $K_{ij} = \sum_{s=1}^{N} \exp(-\gamma_{is}t')\sigma_{sj}(\infty)$, for $t' > 0$. For the one-dimensional case in equation 4.51 a positive γ yields a stationary solution (van Kampen 2007).
The Fokker-Planck equation corresponding to the equation 4.51 reads (Risken 1989):

$$\frac{\partial P}{\partial t} = \sum_{i,j=1}^{N} \gamma_{ij}\frac{\partial}{\partial \psi_i}(\psi_j P) + \sum_{i,j=1}^{N} D_{ij}\frac{\partial^2}{\partial \psi_i \partial \psi_j}P. \tag{4.59}$$

Due to Risken, the Fokker-Planck equation describing an Ornstein-Uhlenbeck process can be solved exactly by a Gaussian distribution which is fully determined by the first and second moments whereby initial values or initial distributions have to be taken into account.

4.2.3.3 Colored noise in a Fokker-Planck equation

The mathematical description for a composite Markov process is taken from (van Kampen 2007). For a Langevin equation with a colored noise as equation 3.6, the composite Markov process means that the behavior of the one variable space can be decomposed from the other variable space. The result is that the contributions from both variables can be added separately in the Fokker-Planck equation. Corresponding to the two coupled Langevin equations 3.6 and 3.8 which describe a colored-noise

process, a Fokker-Planck equation can be derived as follows (van Kampen 2007): In case that the colored noise can be modeled as an Ornstein-Uhlenbeck process,

$$\frac{\partial P(h,\psi,t)}{\partial t} = -\frac{\partial}{\partial h}\Big((A(h)+C(h)\psi(t))P\Big) + \frac{1}{\tau_{corr}}\frac{\partial}{\partial \psi}(\psi P) + \frac{\Gamma_L}{2}\frac{\partial^2 P}{\partial \psi^2}, \tag{4.60}$$

with Γ_L as a diffusion coefficient due to the white noise ξ in equation 3.8.

4.3 Application to Brownian particles

For Brownian particles, a strict differentiation between the Fokker-Planck equation in position space and the Fokker-Planck equation in velocity space can be made. This is due to the so-called separation of time scales (Dhont 1996), (van Kampen 2007). The result is a separate Fokker-Planck equation for the Brownian position variable, i.e. Smoluchowski equation, and the velocity variable, i.e. Rayleigh equation (van Kampen 2007). Further, position-dependent external force fields may require to derive a variable in coupled position-velocity space, i.e. Kramers equation (van Kampen 2007). The Kramers equation is an important aspect in the present work. In the next chapter, it will be outlined that in the non-Brownian case can be discovered similar conditions (Lukassen & Oberlack 2014b) as those that lead to the Kramers equation in the Brownian case.

4.3.1 The Smoluchowski equation

Starting point is the Liouville equation, an equation for one or N Brownian particles and n solvent molecules in position and velocity space, cf. (Dhont 1996) that has been widely studied, e.g. by Lebowitz & Rubin (1963), Deutch & Oppenheim (1971). Reduced forms have been derived, e.g. for the distribution function of two Brownian particles (Mazo 1969). Murphy & Aguirre (1972) derived the N-particle Fokker-Planck equation in position space, see also (Ermak & McCammon 1978) for more information. The Smoluchowski equation (von Smoluchowski 1916) is the Fokker-Planck equation for Brownian particles in position space on the time scale τ_D (equation 2.17) (Dhont 1996). On τ_D the Brownian velocity variable is uncorrelated.

The following considerations are based on (Dhont 1996) and (van Kampen 2007). The change of the particle position can be calculated for time steps $\triangle t$ on τ_D, where the momentum variable is already relaxed to equilibrium. The position variable is a Markov variable which means that the update of the position variable requires knowledge of the position from the last time step but not from earlier time steps. It is possible to define a time step $\triangle t$ for the integration of the Langevin equation 3.12 such that the velocity is uncorrelated but the position and the configuration is constant over the time step: $\tau_p \ll \triangle t < \tau_D$. That means, $\triangle t$ is on the time scale τ_D but small. The coefficients for the Smoluchowski equation according to the equations 4.44 and 4.45 are

computed from the Langevin equation 3.12 for this $\triangle t$. The one-particle Smoluchowski equation is given through (Risken 1989), (van Kampen 2007):

$$\frac{\partial P}{\partial t} = -\frac{\partial}{\partial x}\left(\frac{F(x)}{fr}P\right) + \frac{kT}{fr}\frac{\partial^2 P}{\partial x^2}. \tag{4.61}$$

$F(x)$ may be a position-dependent force field with the pre-condition that it changes very slowly on τ_D (van Kampen 2007). This pre-condition will be clarified in the next subsection 4.3.2. In principle, equation 4.61 is a quasi-linear equation (van Kampen 2007). $\frac{kT}{fr}$ is the Stokes-Einstein diffusivity introduced in equation 2.17 in section 2.3. The N particle equation, corresponding to the Langevin equation for many hydrodynamically interacting particles in shear flow 3.32, is given by (Brady & Bossis 1988), cf. (Dhont 1996):

$$\frac{\partial P}{\partial t} = -\nabla \cdot (\boldsymbol{U}^{\infty} + \boldsymbol{R}_{FU}^{-1} \cdot (\boldsymbol{R}_{FE} : \boldsymbol{E}^{\infty} + \boldsymbol{F}^{P} - kT\nabla lnP))P. \tag{4.62}$$

The operator ":" first appeared in equation 3.36 and contracts the dimensions of the tensors to a vector (cf. (Dhont 1996)). Especially for dilute suspensions, the pair-distribution function turned out to be an advantageous tool to describe the microstructure (Russel et al. 1991), (Batchelor & Green 1972a). It can be derived from the many-body Fokker-Planck equation for Brownian particles, see also (Brady & Morris 1997), (Morris 2009). The pair-distribution function, also referred to as pair-correlation function, can even be derived such that it contains the influence of the remaining particles (Dhont 1996).

4.3.2 Rayleigh equation and Kramers equation

The Rayleigh equation is a Fokker-Planck equation for Brownian particles in pure velocity space (van Kampen 2007) (cf. (Lord Rayleigh 1891) for one dimension), see also (Hoare 1971) in the context of the so-called *Rayleigh gas*. This section is based on (van Kampen 2007).
The Rayleigh equation arises for regarded time steps $\triangle t$ on the time scale τ_p. That means for time steps small on τ_p, $\triangle t < \tau_p$, the velocity is still correlated. Further, the velocity is a Markov variable on τ_p. The position is constant throughout the time steps. From the equation for the change of velocity of a Brownian particle 3.12 with $\tau_p = m/fr$ follows:

$$\frac{\partial P(U,t)}{\partial t} = \frac{1}{\tau_p}\frac{\partial}{\partial U}(UP) + \frac{1}{\tau_p}\frac{kT}{m}\frac{\partial^2 P}{\partial U^2}, \tag{4.63}$$

$$\text{with: } \frac{\langle \triangle U \rangle}{\triangle t} = -\frac{1}{\tau_p}U \tag{4.64}$$

$$\text{and } \frac{\langle (\triangle U)^2 \rangle}{\triangle t} = \frac{1}{\tau_p}\frac{kT}{m}. \tag{4.65}$$

The Brownian velocity in equilibrium is an Ornstein-Uhlenbeck process, cf. equation 4.59. In equilibrium means that initial velocities are damped out (van Kampen 2007), ensemble averages are not time dependent and also the probability distribution is not time dependent, cf. (Dhont 1996). The stationary distribution follows as:

$$P(U) = \left(\frac{m}{2\pi kT}\right)^{1/2} \exp\left(-\frac{m}{2kT}U^2\right). \tag{4.66}$$

Of course, this equation can also be derived for three dimensions (Risken 1989). The mean square displacement grows quadratically in time on τ_p, see equation 3.24 for $\triangle t = t_{n+1} - t_n \to 0$.

A very special case occurs when the position-dependent force field in equation 4.61 does not vary sufficiently slowly. The following explanations are summarized from (van Kampen 2007). Equation 4.61 is the Fokker-Planck equation on the time scale τ_D. For a too fast changing external force field, the regarded time scale has to be scaled down to τ_p so that $F(x)$ is constant over a time step $\triangle t$ on τ_p. This will be further indicated in section 5.2. The Fokker-Planck equation referring to the time scale τ_p is the Rayleigh equation 4.63 which does not take into account any position dependency. Since the change of velocity on τ_p now depends on a position-dependent force field $F(x)$, the velocity variable is no longer Markovian. The solution is a coupled variable of position and velocity (x, U) which is again a Markov variable (van Kampen 2007). This was also outlined in section 4.2.1. The corresponding Fokker-Planck equation for the coupled variable is called Kramers equation (also sometimes referred to as Klein-Kramers equation) (Kramers 1940), (Klein 1921), (van Kampen 2007):

$$\frac{\partial P(x,U,t)}{\partial t} = -U\frac{\partial P}{\partial x} - \frac{F(x)}{m}\frac{\partial P}{\partial U} + \frac{1}{\tau_p}\frac{\partial}{\partial U}(UP) + \frac{1}{\tau_p}\frac{kT}{m}\frac{\partial^2 P}{\partial U^2}. \tag{4.67}$$

The Kramers equation 4.67 corresponds to the Langevin equation given in equation 3.27. There are no coupled xU-terms in the equation. In calculating the coefficients, with $\langle\triangle x\rangle_{x,U} = U\triangle t$ and $\langle\triangle U\rangle_{x,v} = \left(\frac{F(x)}{m} - \frac{1}{\tau_p}U\right)\triangle t$, only the $\langle(\triangle U)^2\rangle_{x,U}$ is of order $O(\triangle t)$, the other terms are all $O((\triangle t)^2)$ (van Kampen 2007). The coefficients in the Fokker-Planck equation are received by dividing by $\triangle t$, see equation 4.44 and 4.45, for $\triangle t \to 0$ on the respective time scale. That means, $O((\triangle t)^2)$-terms for the mean square variables approach zero. The indices given at the $\langle\rangle$-brackets refer to the dependency on the given values of $x(t)$ and $U(t)$ at the beginning of the time step, see also section 4.2.3 where the indices first appeared in equation 4.44. This equation has also been given in three-dimensional form, e.g. from (Risken 1989):

$$\frac{\partial P}{\partial t} = -U_i\frac{\partial}{\partial x_i}P - \frac{F_i(x)}{m}\frac{\partial P}{\partial U_i} + \frac{1}{\tau_p}\frac{\partial}{\partial U_i}(U_iP) + \frac{1}{\tau_p}\frac{kT}{m}\frac{\partial^2 P}{\partial U_i \partial U_i}. \tag{4.68}$$

4.4 Short summary of chapter 4

The present work focuses on the description of non-Brownian particles in shear-induced diffusion by a differential equation for the probability density of an underlying variable of interest. In second order this is referred to as Fokker-Planck equation. Such a Fokker-Planck equation relies on the determination of a Markov variable. In the Brownian case, the separation of time scales enables to derive separate Fokker-Planck equations for the position and velocity variable (van Kampen 2007). An underlying position-dependent force field may result in a coupled position-velocity Fokker-Planck equation referred to as Kramers equation. Further, the Ornstein-Uhlenbeck process is introduced which is of importance in the derivation of the new velocity model in chapter 6. Chapters 2, 3, and 4 give the fundamentals summed up from literature which are necessary for the derivation of the results of the present work. The results are presented in the following chapters 5, 6, and 7.

5 New multiple time scale approach

The Langevin equation 3.32 as given by (Brady & Bossis 1988) in section 3.2 is the starting point for the theoretical considerations in the present work. From this equation can be derived an equation for the velocity of a non-Brownian particle which is also used in (Sierou & Brady 2004) and (Breedveld et al. 2002). Here, the non-dimensionalized Langevin equation shall be regarded and a new compact form is derived which allows to analyze the Langevin equation on all time scales of interest and especially with regard to the Markov property of the respective variables. This chapter is heavily based on the publication (Lukassen & Oberlack 2014b). Basic considerations about the underlying time scales have also been presented in (Lukassen 2012).

5.1 Introduction to the dimensionless equation

In section 2.3, the three fundamental time scales of the present work were introduced, τ_p, τ_f, and τ_D (equations 2.15-2.17). In addition to that, the successional analysis requires to introduce three further time scales which serve as placeholders:

$$\tau_a, \quad \text{a placeholder for the regarded time scale,} \tag{5.1}$$

$$\tau_{conf}, \quad \text{the time scale on which the particle configuration of the system changes,} \tag{5.2}$$

$$\tau_{ac}, \quad \text{the time scale for the velocity autocorrelation.} \tag{5.3}$$

The system can be analyzed with respect to any of the given time scales τ_p, τ_f, and τ_D by setting τ_a to the time scale of interest. Then, the time t and the time step size $\triangle t$ are taken in relation to τ_a. τ_{conf} is defined such that during a time step $\triangle t$ on τ_{conf}, the characteristic distance a particle has moved is a fraction of its own radius a. The determination of τ_{conf} and τ_{ac} differs significantly from Brownian to non-Brownian particles.
As pointed out in section 2.3.1, Brownian particles exhibit a so-called separation of time scales (see (Ermak & McCammon 1978), (van Kampen 2007), (Murphy & Aguirre 1972), (Subramanian & Brady 2004)). That means, in the Brownian case, the position of the particle changes on a significantly larger time scale than the velocity, so $\tau_{conf} = \tau_D$ and $\tau_{ac} = \tau_p$ with $\tau_D \gg \tau_p$. This separation of time scales enables the definition of a time step $\triangle t$ in the range $\tau_p \ll \triangle t < \tau_D$ for the integration of the Langevin equation 3.32 such that the configuration (and thus $\boldsymbol{R}_{FU}$, $\boldsymbol{R}_{FE}$, and $\boldsymbol{U}^{\infty}$) and forces on the particle, e.g. $\boldsymbol{F}^H$ and $\boldsymbol{F}^P$, effectively stay unchanged over the time step while the random part of the velocity resulting from the Brownian motion is completely uncorrelated with the random part from the previous time step (Ermak & McCammon 1978), (Murphy &

Aguirre 1972).
For non-Brownian particles, see (Sierou & Brady 2004), the diffusive behavior results from hydrodynamic interaction between the particles, thus from the hydrodynamic part of the velocity. In contrast to the Brownian velocity with short autocorrelation times, the hydrodynamic velocity component directly depends on the configuration. Consequently, the time scale of the hydrodynamic velocity correlation is the same as the time scale of position change. As already indicated in section 2.3.2, τ_f is the only appearing time scale in the non-Brownian case, therefore, $\tau_{ac} = \tau_f$ and $\tau_{conf} = \tau_f$. That means, for $\triangle t \to 0$ on $\tau_a = \tau_f$, i.e. $\triangle t < \tau_f$, the present velocity is still correlated with velocities from previous time steps while the configuration is considered as unchanged over the time step. In contrast, for large $\triangle t > \tau_f$, the forces and configuration are not constant over the time step, in return, the velocities are uncorrelated. Note that in accordance with the description in section 2.1.2, the $< \tau_f$ and $> \tau_f$ denote time steps on the time scale τ_f, in contrast to $\ll$ and $\gg$. A summary of all the time scales and phenomena that occur on these time scales can be found in tables 6.1, 6.2 and 6.3.

In addition to the different time scales, the system imbeds different length scales. As τ_{conf} is the time scale on which the configuration and the position change, the appropriate length to non-dimensionalize all lengths on $\tau_a = \tau_{conf}$ is the particle radius a, cf. (Brady & Bossis 1988). The distance, a particle moves in a time step $\triangle t$ on τ_{conf}, is a length comparable to the radius.
The appendant length scale to the regarded time scale $\tau_a = \tau_p$ is the distance the particle moves during τ_p. So the characteristic length scale on τ_p is the correlation length of a Brownian particle with the velocity $\sqrt{\frac{kT}{m}}$, see e.g. (Bakunin 2011):

$$l \approx \sqrt{\frac{kT}{m}}\tau_p, \tag{5.4}$$

in the absence of any other forces which could introduce additional length scales. The equilibrium mean square velocity kT/m has been introduced in equation 3.22 in section 3.1.3.
From $\tau_p \ll \tau_D$ with τ_p from equation 2.15 and τ_D from equation 2.17 it follows $l \ll a$, with a as the particle radius.

In the following, the non-dimensionalized form of the Langevin equation 3.32 is analyzed. Depending on the physical situation, special forms of non-dimensionalizing 3.32 were developed, cf. (Brady & Bossis 1988), (Ermak & McCammon 1978), (Foss & Brady 1999), (Subramanian & Brady 2006), (Dhont 1996). In this context, multiple time scale analysis has been used to receive reduced forms of the Fokker-Planck equation for Brownian particles (Wycoff & Balazs 1987), (Subramanian & Brady 2004). However, in the present work is derived a new form of multiple time and length scale analysis which includes all time and length scales of interest for both Brownian and non-Brownian particles. This will be the starting equation for chapter 6. This new compact form that will be exposed for Brownian and non-Brownian particles shall consolidate the relation between the equations of motions and their corresponding Fokker-Planck equations as it will be outlined which variable is a Markov variable

in the corresponding Fokker-Planck equation. The key aspect in this analysis is to point out that the determination of a Markov variable for non-Brownian particles in shear-induced diffusion is the essential challenge. The above mentioned different time scales for Brownian and non-Brownian particles will become apparent below. The analysis of the Brownian particle case which incorporates the white noise due to Brownian motion is done in order to show the special case of the Kramers equation (see e.g. (van Kampen 2007), section 4.3.2) which poses a fundamental mean for the argumentation of the present work for the non-Brownian case.

For the non-dimensionalization of equation 3.32, the assumptions from (Brady & Bossis 1988) and (Foss & Brady 1999) are employed, which imply that the hydrodynamic resistance matrices $\boldsymbol{R}_{FU}$ and $\boldsymbol{R}_{FE}$, cf. equation 3.36, are respectively non-dimensionalized with the friction coefficient $fr = 6\pi\eta a$ and the product $fr \cdot a$ whereby in the present work the non-dimensionalization of $\boldsymbol{R}_{FE}$ is modified to $fr \cdot h_l$. h_l is the placeholder for the length scales a and l, depending on the respective time scale that is regarded. The asymptotics will show that the component referring to l is negligible, however it is introduced to present the full analysis on both the time scales. The components of $\boldsymbol{E}^{\infty}$, cf. equation 3.37, are non-dimensionalized by $\tau_f = 1/\dot{\gamma}$. The mass m of one particle, is subsequently used for any non-dimensionalization of quantities with mass dimension.

In the following, all components of the Langevin equation 3.32 with respect to all time scales are considered in the dimensionless equation. As a first step, all components are decomposed into their different parts in order to understand the processes on the respective time scales. As already pointed out in section 3.2, only the translational components of equation 3.32 are regarded. There will not be any identification mark or index to point that out. In the following analysis, the subsequent expansion parameters are introduced,

$$\epsilon_1 = \frac{\tau_p}{\tau_D} \quad \text{and} \quad \epsilon_2 = \frac{\tau_p}{\tau_f}, \tag{5.5}$$

i.e. ϵ_1 as a small parameter for the asymptotic analysis of the Brownian case and ϵ_2 as a small parameter for the asymptotic analysis of the non-Brownian case. ϵ_2 equals the Stokes number, cf. equation 2.18.

For a better understanding of the time scale expansion to follow, the key time scales τ_p and τ_{conf} are considered separately for each term in an additive form, whereby equation 5.11 is received by inserting τ_p and τ_D into equation 3.39:

$$\tilde{t}_1 = t/\tau_p, \;\; \tilde{t}_2 = t/\tau_{conf}, \tag{5.6}$$

$$\boldsymbol{x} = l\, \tilde{\boldsymbol{x}}_1(\tilde{t}_1) + a\, \tilde{\boldsymbol{x}}_2(\tilde{t}_2), \tag{5.7}$$

$$\boldsymbol{U} = \frac{d\boldsymbol{x}}{dt} = \frac{l}{\tau_p}\tilde{\boldsymbol{U}}_1(\tilde{t}_1) + \frac{a}{\tau_{conf}}\tilde{\boldsymbol{U}}_2(\tilde{t}_2), \tag{5.8}$$

$$\boldsymbol{U}^{\infty}_{\alpha} = (\dot{\gamma} y_{\alpha}, 0, 0) = \left(\frac{1}{\tau_f}(l\, \tilde{y}_{1\alpha}(\tilde{t}_1) + a\, \tilde{y}_{2\alpha}(\tilde{t}_2)), 0, 0\right), \tag{5.9}$$

$$\boldsymbol{U}^{\infty} = \frac{1}{\tau_f}\left(l\, \tilde{\boldsymbol{U}}^{\infty}_1(\tilde{\boldsymbol{x}}_1) + a\, \tilde{\boldsymbol{U}}^{\infty}_2(\tilde{\boldsymbol{x}}_2)\right), \tag{5.10}$$

$$\boldsymbol{F}^B = \frac{m}{\tau_p}\frac{l}{\tau_p}\tilde{\boldsymbol{F}}^B_1(\tilde{t}_1) + \frac{m}{\tau_p}\frac{a}{\tau_D}\tilde{\boldsymbol{F}}^B_2(\tilde{t}_2). \tag{5.11}$$

The tilde-symbol represents the dimensionless components. The index α represents the α-th particle. The indices 1 and 2 identify the two components of the variables on the respective time scales. The components with index 1 refer to equations on the time scale $\tau_a = \tau_p$ while the components with index 2 refer to equations on $\tau_a = \tau_{conf}$. The interparticle force $\boldsymbol{F}^P$ is set to zero in the present work. Implementing 5.6-5.11 into equation 3.32 where $\boldsymbol{F}^H$ has been replaced by 3.36 and $\boldsymbol{F}^P$ is set to zero, yields:

$$m\left(\frac{l}{\tau_p\tau_p}\frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} + \frac{a}{\tau_{conf}\tau_{conf}}\frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2}\right) = -fr\tilde{\boldsymbol{R}}_{FU}\left(\frac{l}{\tau_p}\tilde{\boldsymbol{U}}_1 + \frac{a}{\tau_{conf}}\tilde{\boldsymbol{U}}_2\right) + fr\tilde{\boldsymbol{R}}_{FU}\left(\frac{l}{\tau_f}\tilde{\boldsymbol{U}}^{\infty}_1 + \frac{a}{\tau_f}\tilde{\boldsymbol{U}}^{\infty}_2\right)$$
$$+fr\frac{l}{\tau_f}(\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_1 + fr\frac{a}{\tau_f}(\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_2 + \frac{ml}{\tau_p\tau_p}\tilde{\boldsymbol{F}}^B_1 + \frac{ma}{\tau_p\tau_D}\tilde{\boldsymbol{F}}^B_2. \tag{5.12}$$

In the following, equation 5.12 is regarded separately for Brownian and non-Brownian particles whereby τ_{conf} is set to the respective time scale.

5.2 Brownian particles

For Brownian particles, $\tau_{conf} = \tau_D$ is inserted into equation 5.12. Furthermore, equation 5.12 is multiplied by $\frac{\tau_p\tau_p}{ma}$. Terms τ_p/τ_f can be set to $Pe\epsilon_1$ by using equations 2.20 and 5.5. After applying equation 5.5 with $\frac{\tau_p}{\tau_f} = Pe\ \epsilon_1$ and $\frac{l}{a} = \sqrt{\frac{\tau_p}{\tau_D}} = \epsilon_1^{1/2}$, the dimensionless form for Brownian particles reads:

$$\left(\epsilon_1^{1/2}\frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} + \epsilon_1^2\frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2}\right) = -\epsilon_1^{1/2}\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_1 - \epsilon_1\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2 + Pe\ \epsilon_1^{3/2}\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}^{\infty}_1 + Pe\ \epsilon_1\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}^{\infty}_2$$
$$+Pe\ \epsilon_1^{3/2}(\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_1 + Pe\ \epsilon_1(\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_2 + \epsilon_1^{1/2}\tilde{\boldsymbol{F}}^B_1 + \epsilon_1\tilde{\boldsymbol{F}}^B_2. \tag{5.13}$$

Ordering 5.13 according to the small parameter $\epsilon_1 = \frac{\tau_p}{\tau_D}$, yields to leading order the asymptotics for the Brownian case in the separate equations 5.14 - 5.16 and 5.19 - 5.20:

$$\epsilon_1^{1/2}\text{-terms: } \frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} = -\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_1 + \tilde{\boldsymbol{F}}_1^B. \tag{5.14}$$

5.14 is an equation of motion on $\tau_a = \tau_p$, so $\triangle t$ is a fraction of τ_p here. As already indicated above, this is the time scale of velocity correlation for Brownian particles, i.e. $\tau_{ac} = \tau_p$. An analog equation in one dimension has been introduced in section 3.1.3, equation 3.12 according to (Zwanzig 2001), (Dhont 1996), cf. also (van Kampen 2007). The corresponding Fokker-Planck equation to equation 5.14, namely Rayleigh equation (van Kampen 2007), has been presented in equation 4.63, which is a Fokker-Planck equation in velocity space, as the velocity is a Markov variable here, while the position is not. Of course, the appendant redimensionalized $\boldsymbol{R}_{FU}$ has to be known. This may cause difficulties as on τ_p it may not be assured that disturbances are transferred instantaneously and the matrix is time dependent, see explanation from (Dhont 1996) in the context of equation 3.34.

$$\epsilon_1\text{-terms: } 0 = -\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2 + Pe\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2^{\infty} + Pe(\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_2 + \tilde{\boldsymbol{F}}_2^B, \tag{5.15}$$

$$\text{thus, for } Pe \to 0\text{: } 0 = -\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2 + \tilde{\boldsymbol{F}}_2^B, \tag{5.16}$$

Equation 5.15 is an equation of motion on $\tau_a = \tau_D$, thus $\triangle t$ is a fraction of τ_D. Isolating $\tilde{\boldsymbol{U}}_2$ in 5.15 by multiplying the inverse of $\tilde{\boldsymbol{R}}_{FU}$ and using $\boldsymbol{U} = \frac{d\boldsymbol{x}}{dt}$ from 5.8 leads to a differential equation in position space, namely an equation for $d\boldsymbol{x}/dt$. One more integration of 5.15 with respect to time yields the change of the particle positions with Péclet number dependency, see dimensional analysis in (Brady & Bossis 1988), (Ermak & McCammon 1978). The position-space differential equation in turn may be reformulated to an N-particle Fokker-Planck equation in position space as the positions of all particles are Markovian, see also (Brady & Bossis 1988), (Ermak & McCammon 1978), see equation 4.62 from (Brady & Bossis 1988), cf. (Dhont 1996). Equation 5.16 is the appendant equation for dominant Brownian motion where the shear flow terms are negligible.

Before proceeding with the successively higher order terms in ϵ_1, in the following may shortly be outlined the effects of a position-dependent external force field $\boldsymbol{F}(\boldsymbol{x})$ added on the left-hand side of the Langevin equation 3.32. This has already been mentioned in section 3.1.3 (equation 3.27) and in section 4.3.2 (equation 4.67). The motivation for discussing this effect is that the present work obeys a similar argumentation for the use of an alternative Fokker-Planck equation for non-Brownian particles. Transferred to the present terminology, such an additional external force $\boldsymbol{F}(\boldsymbol{x})$ could be non-dimensionalized by:

$$\boldsymbol{F}(\boldsymbol{x}) = \frac{m}{\tau_k\tau_b}\left(l\,\tilde{\boldsymbol{F}}_1(\boldsymbol{x}/l) + a\,\tilde{\boldsymbol{F}}_2(\boldsymbol{x}/a)\right) = \frac{m}{\tau_k\tau_b}\left(l\,\tilde{\boldsymbol{F}}_1(\tilde{\boldsymbol{x}}_1) + a\,\tilde{\boldsymbol{F}}_2(\tilde{\boldsymbol{x}}_2)\right), \tag{5.17}$$

while the time scales τ_k and τ_b will subsequently be identified with some of the given time scales τ_p, τ_D, or τ_f. Two cases are distinguished below. In case that τ_k and τ_b do

not equal τ_p, τ_D, or τ_f, additional time scales have to be introduced in the asymptotics including the dimensionless analysis for $\boldsymbol{x}$ (equation 5.7) and $\boldsymbol{U}$ (equation 5.8). The explanations to follow stick to the simpler cases that τ_k and τ_b equal either τ_p or τ_D with $\tau_k\tau_b = \tau_p\tau_D$ or $\tau_k\tau_b = \tau_p^2$. Whereby the case that $\tau_k\tau_b = \tau_p^2$ prohibits the use of the Fokker-Planck equation in position space but necessitates using the Kramers equation (see e.g. (Risken 1989), (van Kampen 2007), (Wilemski 1976), (Wycoff & Balazs 1987)). The Kramers equation was introduced in section 4.3.2.
Wilemski (1976) (among others like e.g. Wycoff & Balazs (1987)) gives a rule for the use of the Fokker-Planck equation in position:

$$\left| \frac{\tau_p^2}{m} \frac{\partial \boldsymbol{F}(\boldsymbol{x})}{\partial x} \right| \ll 1. \tag{5.18}$$

Inserting $\boldsymbol{F}(\boldsymbol{x})$ with $\tau_k\tau_b = \tau_p\tau_D$ into equation 5.18 does not pose any problems as the condition is fulfilled. This refers to van Kampen's argument of a sufficiently slowly varying force field, see section 4.3.2. The corresponding Fokker-Planck equation is an equation in position space, as for equation 5.15, see equation 4.61. For the second case that $\tau_k\tau_b = \tau_p^2$, equation 5.18 is not fulfilled. This means a too fast changing force field (van Kampen 2007). After adding the force $\boldsymbol{F}(\boldsymbol{x})$ from equation 5.17 with $\tau_k\tau_b = \tau_p^2$ to the right hand side of equation 5.12, it is obvious that these terms dominate on the τ_D time scale, i.e. in equation 5.15. This is the reason that the regarded time scale $\tau_a = \tau_D$ has to be scaled down to $\tau_a = \tau_p$ (van Kampen 2007). The emerging equation of interest for this case is the equation of motion on the smaller time scale τ_p, i.e. equation 5.14, respectively equation 3.12. The corresponding Fokker-Planck equation to 5.14 is the Rayleigh equation, i.e. a Fokker-Planck equation in velocity space, which does not take into account the position dependency of $\boldsymbol{F}(\boldsymbol{x})$. As the velocity on τ_p is no longer Markovian, the coupled variable of $(\boldsymbol{x}, \boldsymbol{U})$ is used and the resulting Fokker-Planck equation is the Kramers equation 4.67 on the time scale τ_p (van Kampen 2007).
The outstanding ϵ_1-terms of the analysis follow subsequently:

$$Pe\ \epsilon_1^{3/2}\text{-terms: } 0 = \tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_1^{\infty} + (\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_1, \tag{5.19}$$

$$\text{and } \epsilon_1^2\text{-terms: } \frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2} = 0. \tag{5.20}$$

Equation 5.19 is as well as equation 5.14 an equation on τ_p. A comparison of these equations shows that equation 5.19 with $\epsilon_1^{3/2}$-components can be neglected when compared to equation 5.14 with the $\epsilon_1^{1/2}$-components. The same argumentation applies for equation 5.20 when compared to 5.15 which are both equations on τ_D. Equation 5.20 with ϵ_1^2-components can be neglected when compared to equation 5.15 with ϵ_1-components. Thus, in the following, equations 5.19 and 5.20 are not considered.

5.3 Non-Brownian particles

For non-Brownian particles, the configuration changes on τ_f, thus $\tau_c = \tau_f$. Setting this into equation 5.12 above and multiplying by $\frac{\tau_p \tau_p}{ma}$ yields with $\frac{\tau_p}{\tau_D} = \frac{1}{Pe}\frac{\tau_p}{\tau_f}$ and $\frac{l}{a} = \sqrt{\frac{1}{Pe}\frac{\tau_p}{\tau_f}} = \sqrt{\frac{1}{Pe}}\epsilon_2^{1/2}$:

$$\left(\sqrt{\frac{1}{Pe}}\,\epsilon_2^{1/2}\frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} + \epsilon_2^2\frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2}\right) = -\sqrt{\frac{1}{Pe}}\,\epsilon_2^{1/2}\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_1 - \epsilon_2\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2 + \sqrt{\frac{1}{Pe}}\,\epsilon_2^{3/2}\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_1^{\infty}$$
$$+\epsilon_2\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2^{\infty} + \sqrt{\frac{1}{Pe}}\,\epsilon_2^{3/2}(\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_1 + \epsilon_2(\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_2 + \sqrt{\frac{1}{Pe}}\,\epsilon_2^{1/2}\tilde{\boldsymbol{F}}_1^B + \frac{1}{Pe}\,\epsilon_2\tilde{\boldsymbol{F}}_2^B. \tag{5.21}$$

Ordering according to small $\epsilon_2 = \frac{\tau_p}{\tau_f} \ll 1$, (which means a small Stokes number) yields the separate equations 5.22 - 5.26:

$$\sqrt{\frac{1}{Pe}}\,\epsilon_2^{1/2}\text{-terms:}\quad \frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} = -\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_1 + \tilde{\boldsymbol{F}}_1^B. \tag{5.22}$$

Equation 5.22 is the equation of motion on τ_p.
Further,

$$\epsilon_2\text{-terms: } 0 = -\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2 + \tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2^{\infty} + (\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_2 + \frac{1}{Pe}\tilde{\boldsymbol{F}}_2^B, \tag{5.23}$$

$$\text{thus, for } Pe \to \infty\text{: } 0 = -\tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2 + \tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_2^{\infty} + (\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_2. \tag{5.24}$$

Equations 5.23 and 5.24 give the change of position on τ_f. It is important to note that the present approach also covers existing special approaches, as for example equation 5.24 can also be found as part of a Stokes number expansion in $\boldsymbol{U}$ in (Subramanian & Brady 2006).
Finally,

$$\sqrt{\frac{1}{Pe}}\,\epsilon_2^{3/2}\text{-terms: } 0 = \tilde{\boldsymbol{R}}_{FU}\tilde{\boldsymbol{U}}_1^{\infty} + (\tilde{\boldsymbol{R}}_{FE} : \tilde{\boldsymbol{E}}^{\infty})_1, \tag{5.25}$$

$$\text{and } \epsilon_2^2\text{-terms: } \frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2} = 0. \tag{5.26}$$

Besides equation 5.22, equation 5.25 is the second equation on τ_p. Comparing equation 5.22 to 5.25 shows that the shear forces have no influence on τ_p since they are of order $O(\epsilon_2^{3/2})$ in equation 5.25. As $Pe \to \infty$ in the non-Brownian case, equations 5.22 and 5.25 do not have the same significance as the respective equations 5.14 and 5.19 from the Brownian case. As $\tau_{ac} = \tau_f$ and $\tau_{conf} = \tau_f$, the time scale τ_p is not in the scope of interest here. Further, the ϵ_2^2-components of equation 5.26 can be neglected in comparison to the ϵ_2-components of equation 5.23.
As the present work focuses on investigating the Fokker-Planck equation for non-Brownian particles for the case of shear-induced diffusion which occurs on τ_f, equation

5.24 is analyzed in the following. After returning to the dimensional representation and rearranging terms in equation 5.24, it follows for a single particle α:

$$0 = -fr(\boldsymbol{U}_\alpha - \boldsymbol{U}_\alpha^\infty) + \boldsymbol{F}_\alpha^{op} \tag{5.27}$$

$$\text{with} \quad \boldsymbol{F}_\alpha^{op} = -(\boldsymbol{R}_{FU}^*(\boldsymbol{U} - \boldsymbol{U}^\infty))_\alpha + (\boldsymbol{R}_{FE} : \boldsymbol{E}^\infty)_\alpha, \tag{5.28}$$

where $*$ indicates the hydrodynamic influence of all the other particles on a particle α resulting from $-\boldsymbol{R}_{FU}(\boldsymbol{U} - \boldsymbol{U}^\infty)$ in equation 5.24. The α-components are still bold, as they denote a vector with the 3 components for x-, y-, and z-direction. $\boldsymbol{R}_{FU}^*$ still contains non-zero diagonal elements because as indicated in section 3.2 the diagonal components of $\boldsymbol{R}_{FU}$ include both, the friction fr of an isolated particle and added the hydrodynamic influence of the other particles (Dhont 1996). Therefore, the decomposition according to 5.27 is possible with non-zero diagonal elements of $\boldsymbol{R}_{FU}^*$.

In (Breedveld et al. 2002), it is presented an analog to equation 5.27 for the equation of motion of a non-Brownian particle with the hydrodynamic influence of the other particles expressed in a colored-noise force. For the present work, $\boldsymbol{F}_\alpha^{op}$ is also declared as a colored-noise force. Due to the colored-noise force, the equation of motion, cf. 5.27, is called Langevin-*like* equation, cf. (van Kampen 2007).

The starting point for the present work is equation 5.27 which is now rewritten according to:

$$\boldsymbol{U}_\alpha = \boldsymbol{U}_\alpha^\infty + \boldsymbol{V}_\alpha \;\text{ with }\; \boldsymbol{V}_\alpha = \frac{1}{fr}\boldsymbol{F}_\alpha^{op}. \tag{5.29}$$

$\frac{1}{fr}\boldsymbol{F}_\alpha^{op}$ is modeled as a colored-noise velocity $\boldsymbol{V}_\alpha$ in the next chapter 6. Sierou & Brady (2004) have also used a decomposition of the velocity in a shear velocity part and a hydrodynamic part whereby they do not introduce any model for the hydrodynamic velocity. In contrast to equation 3.32 which is an N-particle system of coupled equations, 5.29 now belongs to a system of 3 N independent equations where the velocity $\boldsymbol{V}$ has to be modeled separately.
It will be pointed out below that the derivation of a Fokker-Planck equation strongly depends on the appropriate choice of the regarded time scale τ_a and the time step size $\triangle t$ which is an essential issue of the present work. Most important, a possible alternative approach for the Fokker-Planck equation based on 5.29 is derived.

5.4 Short summary of chapter 5

This chapter includes a time scale analysis which is started by a single equation for both, Brownian and non-Brownian particles. The respective equations on the different time scales have also been presented for separate physical situations in literature. Here, an overall analysis is given which substantiates the equation of motion especially for non-Brownian particles and serves as basis for the analysis of the Markov property of the variables with respect to different time scales.

The Markov property for variables in the Brownian case has widely been analyzed in literature. In contrast to that, the Markov assumption has not been considered in the derivation of the Fokker-Planck equation for non-Brownian particles so far. Due to that, a new compact form of the equation of motion is set up which serves as starting point for both, the Brownian and the non-Brownian case, for the analysis of the Markov property of the underlying variables. The Markov analysis for the Brownian case has been transferred to the present nomenclature to build the basis for the analysis for the non-Brownian case. Further, the overall analysis given here substantiates the equation of motion for non-Brownian particles which serves as basis for the analysis of the Markov property of the variables with respect to different time scales. Such an analysis has not been performed in the context of non-Brownian particles so far.

6 New colored-noise Fokker-Planck equation

In the present chapter, the new colored-noise Fokker-Planck equation will be derived. This equation is set up in coupled colored-noise velocity and position space and is created in order to fulfill the Markov property. The present chapter is heavily based on the author's publication (Lukassen & Oberlack 2014b). The resulting Fokker-Planck equation and first considerations about the derivation were also shown in the author's Master thesis (Lukassen 2012).

6.1 Problems in deriving the Fokker-Planck equation for non-Brownian particles in shear flow

As indicated in the previous sections, non-Brownian particles do not exhibit a separation of time scales (Subramanian & Brady 2004). Further, non-Brownian particles only show long-time diffusive behavior (Sierou & Brady 2004) in contrast to Brownian particles which show both, short- and long-time behavior. The Markov property enables to derive a Fokker-Planck equation valid for long times t on the regarded time scale τ_a based on the behavior of the variables during a small time step $\triangle t$ on τ_a. That means, irrespective of the total time t that shall be regarded in the probability density, the basis is always a small time step $\triangle t$ that needs all information necessary to capture the long time behavior, see (van Kampen 2007). Concerning the problem of non-Brownian particles, the time step size can be chosen large on τ_f where the diffusive regime has started and the velocity is uncorrelated, with the drawback that the configuration changes during the time step. On the other hand, the time step can be chosen small on τ_f where the configuration can be considered as constant during the time step, but the velocity is still correlated. The first way corresponds to the problem of a too fast changing position-dependent force field for Brownian particles which resulted in the derivation of the Kramers equation, i.e. the time scale had to be scaled down (van Kampen 2007). Otherwise, the update of the position variable would require knowledge about how the force field changed during the time step. The shear velocity $\boldsymbol{U}^\infty$ is comparable to the external force field. For the latter way the position is not a Markov variable anymore because the hydrodynamic velocity component is not fully relaxed. This problem shall be solved within this chapter. In the next section however, will be outlined how non-Brownian particles have been described so far.

6.1.1 Literature overview on the Fokker-Planck equation for non-Brownian particles

Non-Brownian particles in shear-induced diffusion have been derived in pure position space only using modified diffusion tensors. The analysis from Santamaría-Holek, Barrios & Rubi (2009a), (Santamaría-Holek et al. 2009b) starts with a coupled Fokker-Planck equation in velocity and position space. They assume that this coupled Fokker-Planck equation can be transformed into a Fokker-Planck equation in position space whereby the diffusion tensor is modified incorporating thermal and non-thermal effects under consideration of the second law of thermodynamcis. In this context, they find a breaking of the fluctuation-dissipation theorem due to an introduced shear flow which also has been investigated e.g. by Mauri & Leporini (2006).

Sierou & Brady (2004) construct a position-space Fokker-Planck equation as follows:

$$\frac{\partial P}{\partial t} = -y\dot{\gamma}\frac{\partial P}{\partial x} + D_{xx}(t)\frac{\partial^2 P}{\partial x^2} + D_{yy}(t)\frac{\partial^2 P}{\partial y^2} + D_{zz}(t)\frac{\partial^2 P}{\partial z^2} + 2D_{xy}(t)\frac{\partial^2 P}{\partial x \partial y}. \tag{6.1}$$

Sierou and Brady point out that the $D_{ij}(t)$ coefficients ($i, j = x, y, z$) in their equation are time dependent diffusion coefficients for the small and intermediate time scales which become constant for long times. Sierou and Brady derive their diffusion coefficients based on the equations 2.28-2.31 (cf. (Elrick 1962)) from section 2.3.3.2. Extracting D_{yy} and D_{zz} from equations 2.28 and 2.29 poses no problem, and for y- and z-direction applies (Sierou & Brady 2004):

$$D_{yy}(t) = \frac{1}{2}\frac{d}{dt}\langle yy\rangle, \tag{6.2}$$

$$D_{zz}(t) = \frac{1}{2}\frac{d}{dt}\langle zz\rangle. \tag{6.3}$$

But extracting D_{xx} and D_{xy} via the mean square displacements in equations 2.30 and 2.31 in analogy to D_{yy} and D_{zz} causes a difficulty, as Sierou and Brady point out, because for very large times t in the non-Brownian case, the terms of order $O(t^2)$ and $O(t^3)$ dominate. So instead, they derive D_{xx} and D_{xy} by calculating $\langle xx\rangle$ and $\langle xy\rangle$ from $\int_0^t \boldsymbol{U}_\alpha(t')dt' = \int_0^t \boldsymbol{U}_\alpha^\infty(t') + \boldsymbol{U}_\alpha^h(t')dt' = \boldsymbol{x}^\infty + \boldsymbol{x}^h$, with $\boldsymbol{x}^h$ as the hydrodynamic displacement due to the hydrodynamic velocity component $\boldsymbol{U}^h$, and the position in the origin for $t = 0$. Then, they use the following relation (resulting from a time derivative of equations 2.30 and 2.31) which also can be found in general form in (van Kampen 2007):

$$\partial_t\langle xx\rangle - 2\langle x\,(\dot{\gamma}y)\rangle = 2D_{xx} \tag{6.4}$$

$$\partial_t\langle xy\rangle - \langle y\,(\dot{\gamma}y)\rangle = 2D_{xy}. \tag{6.5}$$

The resulting diffusion coefficients include a coupling term (Sierou & Brady 2004):

$$D_{xx}(t) = \frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle + \left\langle \frac{dx^h}{dt}\int_0^t \dot{\gamma}y(t')dt' \right\rangle, \tag{6.6}$$

$$D_{xy}(t) = \frac{1}{2}\frac{d}{dt}\langle x^h y\rangle + \frac{1}{2}\left\langle \frac{dy}{dt}\int_0^t \dot{\gamma}y(t')dt' \right\rangle. \tag{6.7}$$

All diffusion coefficients depend on time t. Due to Sierou & Brady (2004), this allows them to use the coefficients also for short times t where the diffusive behavior has not started yet and they claim that the diffusion coefficients become constant for large times t. Still, following the argumentation in the previous section, at arbitrary times t, even in the long-time limit $t \to \infty$, i.e. $t > \tau_f$, the behavior of position is non-diffusive when small time step sizes $\triangle t < \tau_f$ on the time scale $\tau_a = \tau_f$ are used. Consequently, the description of the diffusive regime necessitates time step sizes $\triangle t > \tau_f$. On the other hand, large time step sizes $\triangle t > \tau_f$ cause a problem as will be pointed out in the next section 6.2.

The argumentation in the present work will be similar to problems that arise in the Brownian case, either, when an external force field $\boldsymbol{F}(\boldsymbol{x})$ changes too fast (see section 4.3.2, Kramers equation), or when $\triangle t > \tau_D$. The intention here is not to increase the time step size to $\triangle t > \tau_f$, until the position is diffusive. In contrast, the colored-noise velocity $\boldsymbol{V}_\alpha$ in equation 5.29 is modeled such, that this is diffusive for $\triangle t < \tau_f$ on $\tau_a = \tau_f$. Here, an Ornstein-Uhlenbeck process is used. Ornstein-Uhlenbeck processes with exponential autocorrelation functions are a common scheme to describe colored-noise variables (van Kampen 2007). The principle of velocity autocorrelation functions decaying exponentially in time has been observed in several particle-type situations as e.g. sedimenting non-Brownian particles (Nicolai, Herzhaft, Hinch, Oger & Guazzelli 1995). The mean square of $\boldsymbol{V}_\alpha$ will be linear in time for times $t < \tau_f$. For times $t \to \infty$, $\langle \boldsymbol{V}^2\rangle$ is a constant, in analogy to the Brownian velocity in equilibrium $\langle (U_i(t \to \infty))^2\rangle$ as shown in equation 3.22. A summary on the time scales can be found in tables 6.1,6.2, and 6.3. Further, the colored-noise description requires diffusion coefficients which will be derived from the D_{xx}, D_{yy}, D_{zz}, and D_{xy} via the mean square displacements, cf. equations 2.28-2.31.

Breedveld et al. (1998) use the Fokker-Planck equation only in position space and restrict their equation to the case of long enough time intervals. They measure the shear-induced self-diffusivity by a new correlation technique. Their Fokker-Planck equation has the same form as equation 6.1 but with constant diffusion coefficients.

Later, in (Breedveld et al. 2002), some of the authors from (Breedveld et al. 1998) and co-workers also noticed that a Fokker-Planck equation in position space as given in equation 6.1 may not be suitable, but they did not further analyze this problem as done in the present work. Instead, they derive a Langevin equation with a colored-noise force, which in the mean is zero and contains the hydrodynamic influence of the particles onto each other. With this equation, they also aim at deriving the full position diffusion tensor in a modified form, with a colored-noise force in the equation of motion and thus overcome the common white noise assumption. Nevertheless, they

do not derive a Fokker-Planck equation in a coupled variable based on their Langevin equation.

From mathematics, in literature can also be found the general case of a colored-noise Langevin equation, where a Fokker-Planck-like equation in position space has been derived with a time dependent diffusion coefficient (Hänggi & Jung 1995). However, this has not been used in the context of shear-induced diffusion so far. This will be referred to again in sections 7.1.1 and 8.2.1. Even though, Sierou & Brady (2004) also have a time dependent $D(t)$ they do not give an explicit model for their diffusion coefficients. Their $D(t)$ is based on measuring mean square displacements by numerical simulations. When Sierou and Brady set their diffusion coefficients to constants for long enough times, the information about non-separated time scales and long correlation times is lost. Due to this, in the present work is concluded that the traditional position-space models may be insufficient.

6.2 Colored-noise approach

Here, equation 5.29 is the basis for the derivation of the Fokker-Planck equation of the shear-induced self-diffusion process. From now on, the dependence on time t is written explicitly to account for the correlation of varying times. A one-particle formulation is regarded in the following. The hydrodynamic velocity $V_i(t)$ results from the hydrodynamic interaction on the regarded particle due to all other particles. Here, $V_i(t)$ exhibits a colored-noise property:

$$U_x(t) = U_x^{\infty}(t) + V_x(t) \tag{6.8}$$

$$U_y(t) = V_y(t) \tag{6.9}$$

$$U_z(t) = V_z(t), \tag{6.10}$$

for time $t > \tau_f$ in order to map the diffusive regime. Regarding times $t > \tau_f$ does not mean that $\triangle t > \tau_f$ as will be pointed out below. Note that this is the equation for one particle α in the full system. U_x^{∞} is the first component of $\boldsymbol{U}^{\infty}$, i.e. $\dot{\gamma}y$. The whole system for all N particles consists of 3 N equations. Since the influence of the particles onto each other is going to be modeled separately in $\boldsymbol{V}$, these 3 N equations all are independent. Thus it makes no difference, if the averaging procedures are for a single particle over different configurations, i.e. ensemble averages, cf. section 4.1.4, or if averaging procedures are performed over all particles in one system. Here will be used the latter case for the purpose of manageability. The α-index will be skipped in the following. The essential challenge is to find the corresponding Fokker-Planck equation to equations 6.8-6.10. This implies that it has to be determined which time scale shall be taken as underlying time scale τ_a and which variable is a Markov variable on this time scale.
As shown above in the non-Brownian case, on $\tau_a = \tau_f$, for $\triangle t > \tau_f$ the position is diffusive and as the velocity changes with the configuration, the velocity is uncorrelated for such a time step size. However, the configuration will only be constant for $\triangle t \to 0$,

respectively $\triangle t < \tau_f$. In the following will be determined whether $\triangle t$ should be larger or smaller than τ_f. In a first step, a model for V_i is developed. In a second step, the drift and diffusion terms in position for both, $\triangle t < \tau_f$ and $\triangle t > \tau_f$, will be considered separately to show that time step sizes $\triangle t < \tau_f$ are necessary.

For the derivation of a model for the colored-noise velocity in the present work, a heuristic approach as defined in section 3.1.1 following (Risken 1989) is employed. Presently, it is assumed that the colored-noise velocity $V_i(t)$ itself is a Markov process and further that it can be modeled by an Ornstein-Uhlenbeck process (cf. (Uhlenbeck & Ornstein 1930), (van Kampen 2007), cf. section 4.2.3.2) represented by the following Langevin equation:

$$\frac{dV_i(t)}{dt} = -\frac{1}{\tau_c} V_i(t) + L_i(t), \tag{6.11}$$

for $i = x, y, z$ with white noise $L_i(t)$. τ_c is the correlation time of the colored-noise velocity. Equation 6.11 is a linear Langevin equation which according to van Kampen (2007) does not pose problems in determining the corresponding Fokker-Planck equation, cf. section 3.1.2. The approach follows the Langevin-approach from (van Kampen 2007), cf. section 3.1.1. The denotation as a colored-noise velocity also originates from the fact that this velocity component is correlated on the same time scale as the configuration changes. This poses a strong contrast to the velocity in the Brownian case, which is correlated on τ_p and thus on a much smaller time scale than the change of position. Due to the Markov property, the value of $V_i(t)$ depends on the value from the previous time step but not from earlier time steps. It is important to note that $\boldsymbol{V}$ is defined on τ_f and not on τ_p as it results from the equation of motion 5.27 for non-Brownian particles on τ_f. The new definition of $V_i(t)$ with a time derivative is not related to the time derivative of the velocity on τ_f which is zero in equation 5.25. This colored-noise velocity would have to be embedded in the equation of motion from the beginning. According to Sierou & Brady (2004) the correlation time of the velocity in the non-Brownian case is τ_f. Since in the present colored-noise velocity model (6.11) these long correlation times have to be considered, τ_c is set to τ_f. The white noise $L_i(t)$ introduces a randomness which results in the diffusive behavior in the position space for times $t > \tau_f$, $t \to \infty$.
For an Ornstein-Uhlenbeck process corresponding to equation 6.11, the following results apply in the limit $t \to \infty$ (Risken 1989), (Uhlenbeck & Ornstein 1930), (van Kampen 2007):

$$\langle L_i(t+t')L_j(t)\rangle = B_{ij}\delta(t'), \tag{6.12}$$

$$\langle V_i(t+t')V_j(t)\rangle = \frac{B_{ij}\tau_c}{2}\exp\left(\frac{-|t'|}{\tau_c}\right), \tag{6.13}$$

$$\langle V_i(t)\rangle = 0. \tag{6.14}$$

Since here, the correlation time of $\boldsymbol{V}$ is set to τ_f, equations 6.12-6.14 are rewritten in the following form:

$$\langle L_i(t+t')L_j(t)\rangle = B_{ij}\delta(t'), \tag{6.15}$$

$$\langle V_i(t+t')V_j(t)\rangle = \frac{B_{ij}\tau_f}{2}\exp\left(\frac{-|t'|}{\tau_f}\right), \tag{6.16}$$

$$\langle V_i(t)\rangle = 0. \tag{6.17}$$

$\boldsymbol{V}$ as resulting from equation 6.11 fulfills these equations for $t \to \infty$, which is the equilibrium state, cf. also the Brownian velocity in equilibrium in equation 3.17. The large time t guarantees the independence from the initial value $V_i(0)$. Further, the large time t guarantees to be in the regime where shear-induced diffusion has already started. The exponential term in equation 6.16 accounts for the long correlation times which result in colored noise in contrast to the white noise from the Brownian force $\boldsymbol{F}^B$, cf. equation 3.39. In the next sections, the drift and diffusion terms for the new Fokker-Planck equation are derived based on the Ornstein-Uhlenbeck model for $V(t)$. The derivation of the B_{ij} coefficients will be shown in section 6.3. These B_{ij} coefficients will be the diffusion coefficients in the new colored-noise Fokker-Planck equation.

6.2.1 Drift and diffusion terms for position variables

In the following, the drift and diffusion terms in position space are analyzed with respect to the time step size $\triangle t$ with the result that a time step $\triangle t < \tau_f$ is necessary. The present derivation for the drift and diffusion terms will be done for x-direction only, as this is also the direction of shear flow. y- and z-direction work analogously. The drift term (cf. equation 4.44) for x-direction consists of a term due to the shear flow $\triangle x^\infty$ and a term due to the hydrodynamic interactions $\triangle x^h$ (the rules for these coefficients from e.g. (Risken 1989) and (van Kampen 2007) are used). From equation 6.8 and 6.11 it follows:

$$\langle \triangle x\rangle = \langle \triangle x^\infty\rangle + \langle \triangle x^h\rangle \tag{6.18}$$

$$\text{with} \quad \langle \triangle x^\infty\rangle = \Big\langle \int_{t_n}^{t_{n+1}} y(t)\dot{\gamma}dt\Big\rangle, \tag{6.19}$$

$$\langle \triangle x^h\rangle = \int_{t_n}^{t_{n+1}} \langle V_x(t)\rangle dt, \tag{6.20}$$

$$\text{further,} \quad \langle V_x(t_{n+1})\rangle = \langle V_x(t_n)\rangle \exp\left(-\frac{\triangle t}{\tau_f}\right). \tag{6.21}$$

A similar equation to 6.18 has also been used by Sierou & Brady (2004), without a model for the hydrodynamic velocity. The order of the averaging and the integration can be interchanged, cf. (Haken 1977). Equation 6.19 and 6.20 are regarded separately for both time step sizes, $\triangle t > \tau_f$ and $\triangle t < \tau_f$.

In equation 6.19, y is expanded in a Taylor series around t_n, cf. the derivation of the Fokker-Planck equation in (Haken 1977), (van Kampen 2007):

$$\begin{aligned}\langle \triangle x^\infty \rangle &= \Big\langle \int_{t_n}^{t_{n+1}} y(t_n)\dot{\gamma} dt \Big\rangle + \Big\langle \int_{t_n}^{t_{n+1}} (t-t_n)\frac{dy(t_n)}{dt}\dot{\gamma} dt \Big\rangle + \Big\langle \int_{t_n}^{t_{n+1}} O((t-t_n)^2) dt \Big\rangle \\ &= \langle y(t_n)\rangle \dot{\gamma}\triangle t + \left[\frac{(t-t_n)^2}{2}\Big\langle \frac{dy(t_n)}{dt}\Big\rangle \dot{\gamma}\right]_{t_n}^{t_{n+1}} + O((\triangle t)^3) \\ \frac{\langle \triangle x^\infty \rangle_{\boldsymbol{x},\boldsymbol{V}}}{\triangle t} &= \dot{\gamma}\left(y(t_n) + \frac{\triangle t}{2}\frac{dy(t_n)}{dt}\right) + O((\triangle t)^2). \end{aligned} \tag{6.22}$$

Note, that the $\boldsymbol{x}, \boldsymbol{V}$-index at the $\langle\rangle$-brackets is added in equation 6.22 to account for the conditional average from equation 4.44 with constant $\boldsymbol{x}, \boldsymbol{V}$ at time t_n. Due to that, the $\langle\rangle$-brackets for the constant values at t_n can be removed. Since the only appearing time scale for non-Brownian particles in shear flow is τ_f, τ_a is set to τ_f and thus only the components on τ_f are regarded (and not possible components on τ_p). The major question is if $\triangle t$ should be smaller or larger than τ_f. In the following, the dimensionless form is regarded:

$$\frac{a}{\tau_f}\frac{\langle \triangle \tilde{x}^\infty \rangle_{\boldsymbol{x},\boldsymbol{V}}}{\triangle \tilde{t}} = \frac{1}{\tau_f}\left(a\,\tilde{y}(t_n) + \frac{\tau_f \triangle \tilde{t}}{2}\frac{a}{\tau_f}\frac{d\tilde{y}(t_n)}{d\tilde{t}} + ...\right) \tag{6.23}$$

with $\triangle t = \tau_f\,\triangle \tilde{t}$, $\frac{dy}{dt} = \frac{a}{\tau_f}\frac{d\tilde{y}}{d\tilde{t}}$ (see equation 5.8) and $\dot{\gamma} = \frac{1}{\tau_f}$. Note, that $\frac{dy(t_n)}{dt} = V_y(t_n)$, see equation 6.9. For $\triangle t \to 0$, i.e. $\triangle t < \tau_f$, terms of order $O(\triangle \tilde{t})$ can be neglected since $\triangle \tilde{t} \to 0$. For $\triangle t > \tau_f$, the higher order terms can not be neglected since $\triangle \tilde{t} > 1$.
Consequently for $\triangle t > \tau_f$, the update of the position cannot be realized with the knowledge of only the position of the last time step. $\boldsymbol{U}^\infty$, depending also on the position, cannot be taken as constant over the time step. The conclusion is that, for time steps $\triangle t > \tau_f$, a Fokker-Planck equation in the position is not valid here as the drift term $\boldsymbol{U}^\infty$ (corresponding to an inhomogeneous force field) changes too fast for this time step size, cf. (Kramers equation and equation 5.18).

For the $\langle \triangle x^h \rangle$-part (equation 6.20), it follows for $\triangle t < \tau_f$, $\triangle t \to 0$, cf. equation 6.21:

$$\langle V_x(t_{n+1})\rangle_{\boldsymbol{V}} = \langle V_x(t_n)\rangle_{\boldsymbol{V}} + O(\triangle t), \tag{6.24}$$

$$\langle \triangle x^h \rangle = \int_{t_n}^{t_{n+1}} \langle V_x(t)\rangle dt = \langle V_x(t_n)\rangle \triangle t + O((\triangle t)^2), \tag{6.25}$$

$$\frac{\langle \triangle x^h \rangle_{\boldsymbol{x},\boldsymbol{V}}}{\triangle t} = V_x(t_n) + O(\triangle t), \tag{6.26}$$

where terms of order $O(\triangle t)$ in equation 6.26 can be neglected. The $\boldsymbol{V}$-index refers to the conditional average as above. The $O(\triangle t)$-terms in 6.24 come from the series representation of the exponential function in 6.21.
The contrary case of a $\langle \triangle x^h \rangle$ for $\triangle t > \tau_f$ shall be analyzed for the purpose of completeness. From equation 6.21 follows, that $\langle V_x(t_{n+1})\rangle_{\boldsymbol{V}} \approx 0$ for $\triangle t \to \infty$. The time step

size is longer than the correlation length of the colored-noise velocity and thus using equation 6.21:

$$\langle \triangle x^h \rangle = \int_{t_n}^{t_{n+1}} \langle V_x(t) \rangle dt \tag{6.27}$$

$$= \int_{t_n}^{t_{n+1}} \langle V_x(t_n) \rangle \exp\left(-\frac{(t-t_n)}{\tau_f}\right) dt, \tag{6.28}$$

$$\langle \triangle x^h \rangle_{\boldsymbol{x},\boldsymbol{V}} = \langle V_x(t_n) \rangle_{\boldsymbol{V}} \, \tau_f = V_x(t_n)\tau_f \tag{6.29}$$

$$\text{and thus} \quad \frac{\langle \triangle x^h \rangle_{\boldsymbol{x},\boldsymbol{V}}}{\triangle t} \approx 0. \tag{6.30}$$

Due to the problems arising for large $\triangle t$ in the context of $\boldsymbol{U}^\infty$, in the present work, the time step $\triangle t$ is considered to be much smaller than the time scale of configurational changes, τ_f. For this case, the bulk velocity $\boldsymbol{U}^\infty$ can be considered as constant over the time step. The drift term for the position in the alternative Fokker-Planck equation is achieved by combining equation 6.22 and 6.26 to $\frac{\langle \triangle x \rangle_{\boldsymbol{x},\boldsymbol{V}}}{\triangle t} = \dot{\gamma} y(t_n) + V_x(t_n)$. Hence, the time step size $\triangle t$ employed in the present work is smaller than the correlation time τ_f of V_i.
This colored-noise property destroys the Markov property of the position (for a mathematical substantiation, see e.g. (van Kampen 2007)). Consequently, also for the small time step sizes it is not possible to derive a consistent Fokker-Planck equation in position space.

The analysis for the diffusion terms works analogously. Again, the rules for the diffusion terms for Ornstein-Uhlenbeck processes and Fokker-Planck equations can be found e.g. in (Dhont 1996), (Risken 1989), or (van Kampen 2007). The diffusion coefficients can be achieved by using the following equation (already mentioned in section 4.2.3):

$$\langle \triangle x \triangle x \rangle = \left\langle \left(\int_{t_n}^{t_{n+1}} y(t)\dot{\gamma} dt \right)^2 \right\rangle + 2 \left\langle \int_{t_n}^{t_{n+1}} \int_{t_n}^{t_{n+1}} U_x^\infty(t') V_x(t'') dt' dt'' \right\rangle$$
$$+ \left\langle \int_{t_n}^{t_{n+1}} \int_{t_n}^{t_{n+1}} V_x(t') V_x(t'') dt' dt'' \right\rangle. \tag{6.31}$$

For $\triangle t \to 0$, i.e. $\triangle t < \tau_f$:

$$\langle \triangle x \triangle x \rangle_{\boldsymbol{x},\boldsymbol{V}} = \langle U_x^\infty(t_n)^2 \rangle_{\boldsymbol{x},\boldsymbol{V}} (\triangle t)^2 + 2 \langle U_x^\infty(t_n) V_x(t_n) \rangle_{\boldsymbol{x},\boldsymbol{V}} (\triangle t)^2 + \langle V_x^2(t_n) \rangle_{\boldsymbol{V}} (\triangle t)^2 = 0, \tag{6.32}$$

with $\langle V_x^2(t_n) \rangle_{\boldsymbol{V}} = V_x^2(t_n)$, see also equation 3.24 in section 3.1.3. Equation 6.32 shows the quadratic component that arises for small times (compare simulation results from Sierou & Brady (2004), mentioned above). This is in analogy to the Brownian case for the τ_p time scale. Also in the non-Brownian case, the quadratic behavior implies that the position is not diffusive for $\triangle t \to 0$.
The calculation of the first and second part of equation 6.31 poses further difficulties. But at least for the diffusivity of the position in y- and z-direction (without terms as the first and second part of equation 6.31) it is apparent, that the time step size should

be larger than τ_f in order to reach the linear regime. For $\triangle t \to \infty$, i.e. $\triangle t > \tau_f$, in appendix A in equation A.1 is shown:

$$\langle \triangle x^h \triangle x^h \rangle_{\boldsymbol{x},\boldsymbol{V}} = V_x^2(t_n)\tau_f^2 + B_{xx}\tau_f^2 \triangle t - \frac{3}{2} B_{xx}\tau_f^3, \tag{6.33}$$

thus linear behavior in $\triangle t$.

6.2.2 Drift and diffusion terms for velocity variables

Due to the conflict concerning the appropriate time step size and the Markov process assumption, the traditional Fokker-Planck equation in position space has to be extended. The drift for the colored-noise velocity results from equation 4.44 by calculating $\triangle V_i = V_i(t_{n+1}) - V_i(t_n)$ with equation 6.21 for small time step sizes $\triangle t$.
For the rules for diffusion terms, see again e.g. (Dhont 1996), (Risken 1989), or (van Kampen 2007). On the time scale $\tau_a = \tau_f$ with $\triangle t < \tau_f$, the colored-noise velocity is a diffusive process. This is analog to the Brownian case, where for $\tau_a = \tau_p$ not the position but the velocity is assumed to be diffusive. In the present work, not the whole velocity is assumed to be diffusive but only the hydrodynamic component V_i. It follows:

$$\text{for } \triangle t < \tau_f \text{ and } \triangle t \to 0: \ \langle \triangle V_i \triangle V_j \rangle_{\boldsymbol{V}} = B_{ij} \triangle t \tag{6.34}$$

$$\text{for } \triangle t > \tau_f \text{ and } \triangle t \to \infty: \ \langle \triangle V_i \triangle V_j \rangle_{\boldsymbol{V}} = \frac{B_{ij}\tau_f}{2} + V_i(t_n)V_j(t_n) = const. \tag{6.35}$$

A proof can be found in appendix B and an overview is shown in table 6.3. As already indicated above, in order to avoid the violation of the Markov property of the position variables, a colored-noise approach for the Fokker-Planck equation is used. A general mathematical description for colored-noise problems in one dimension under the assumption of a composite Markov process is given by van Kampen (2007), see section 4.2.3.3. Here, this approach is extended to the three-dimensional shear-induced diffusion problem. This means that by using equation 4.43, there are no diffusion coefficients in position space (as for small time step sizes, the mean square displacements are of order $O((\triangle t)^2)$, cf. equation 6.32) and due to the composite Markov process assumption, no coupled diffusion coefficients of position and the colored-noise velocity. Coupled coefficients would be of order $(\triangle t)^2$ anyway, which is exemplary shown for y-direction here, cf. components of the Kramers equation (van Kampen 2007):

$$\langle \triangle y \triangle V_y \rangle = \left\langle \int_{t_n}^{t_{n+1}} V_y(t')dt' \triangle V_y \right\rangle \tag{6.36}$$

$$= \langle V_y(t_n) \triangle t \triangle V_y \rangle \text{ (see equation 6.25)} \tag{6.37}$$

$$= \langle V_y(t_n)V_y(t_{n+1}) \rangle \triangle t - \langle V_y(t_n)V_y(t_n) \rangle \triangle t \tag{6.38}$$

$$= O((\triangle t)^2) \text{ for } \triangle t \to 0, \tag{6.39}$$

$$\text{with } \triangle V_y = V_y(t_{n+1}) - V_y(t_n). \tag{6.40}$$

In equation 6.39, the autocorrelation from equation 6.16 is expanded in the exponential series. The diffusion coefficients for V_i according to equation 4.45 are obtained from 6.34 as $\frac{\langle \triangle V_i \triangle V_j \rangle_{\boldsymbol{V}}}{\triangle t} = B_{ij}$. Still, the coefficients B_{ij} need to be determined.

Integrating equation 6.16 for $t \to \infty$ yields for B_{ij}:

$$B_{ij} = \int_0^\infty \lim_{t\to\infty} \langle V_i(t+t')V_j(t)\rangle dt' \; \frac{2}{\tau_f} \left(\int_0^\infty \exp\left(\frac{-t'}{\tau_f}\right) dt' \right)^{-1} \tag{6.41}$$

$$= \int_0^\infty \lim_{t\to\infty} \langle V_i(t+t')V_j(t)\rangle dt' \; \frac{2}{\tau_f^2}. \tag{6.42}$$

Consequently, the determination of B_{ij} requires knowledge of $\int_0^\infty \lim_{t\to\infty} \langle V_i(t+t')V_j(t)\rangle dt'$.

The B_{ij} coefficients are not the same as the diffusion coefficients D_{ij}. B_{yy} is connected to D_{yy} using $\langle U_y(t+t')U_y(t)\rangle = \langle V_y(t+t')V_y(t)\rangle$ with equation 6.9.
From the known rule $D_{yy} = \int_0^\infty \lim_{t\to\infty} \langle U_y(t+t')U_y(t)\rangle dt'$ (cf. (Sierou & Brady 2004), (van Kampen 1989), or with a colored-noise force in (Breedveld et al. 2002)) (cf. also Green-Kubo expression e.g. in (Risken 1989)) can be found that

$$D_{yy} = \int_0^\infty \lim_{t\to\infty} \langle V_y(t+t')V_y(t)\rangle dt'. \tag{6.43}$$

For D_{zz} the procedure is analogous.
Equation 6.41 results in:

$$B_{ij} = \frac{2D_{ij}}{\tau_f^2}, \quad \text{for } i,j = y,z. \tag{6.44}$$

A mathematically analog form to equation 6.44 can also be found in (van Kampen 1989). In contrast to D_{ij} which scales as $a^2 \cdot \dot{\gamma} = a^2/\tau_f$, cf. e.g. (Sierou & Brady 2004), the coefficients B_{ij} scale as a^2/τ_f^3, cf. equation 6.44. Note that equation 6.44 applies when setting the correlation time τ_c to τ_f, in general form, the equation would read:

$$B_{ij} = \frac{2D_{ij}}{\tau_c^2}, \quad \text{for } i,j = y,z. \tag{6.45}$$

Under use of the exponentially decaying correlation for long times t, i.e. equation 6.16, for the x-direction applies:

$$\int_0^\infty \lim_{t\to\infty} \langle V_x(t+t')V_x(t)\rangle dt' = D_{xx} - \left\langle \int_0^\infty \lim_{t\to\infty} U^\infty(t')V_x(t)dt' \right\rangle = \frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle, \tag{6.46}$$

for proof, see appendix C. The coupled U^∞, V_x-term corresponds to the coupled term of Sierou & Brady (2004) in equation 6.6. Consequently, it can be obtained:

$$B_{xx} = \frac{2}{\tau_f^2}\frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle. \tag{6.47}$$

For the xy-component, the procedure is analogous (compare proof for the xx-component in appendix C) resulting in:

$$\int_0^\infty \lim_{t\to\infty} \langle V_x(t+t')V_y(t)\rangle dt' = \frac{1}{2}\frac{d}{dt}\langle x^h y\rangle, \tag{6.48}$$

and hence:

$$B_{xy} = \frac{2}{\tau_f^2}\frac{1}{2}\frac{d}{dt}\langle x^h y\rangle. \tag{6.49}$$

Analog to the diffusion coefficients D_{ij} for non-Brownian particles, see section 6.1.1 and 2.3.3.2, (Brady & Morris 1997), (Sierou & Brady 2004), here is assumed that also the B_{ij} coefficients have only the xy-component as non-zero diagonal element. That shows that the B_{ij} coefficients can be derived via the position diffusion coefficients D_{ij}, i.e. the mean square displacements. As already mentioned in the context of the non-Brownian position diffusion tensors D_{ij} in section 2.3.3.2, also the B_{ij}-coefficients do not incorporate any position dependency of the respective particles anymore since these are only 1-particle velocity diffusion tensors. The mean square displacements can be achieved either by experiments, see e.g. the work of Breedveld et al. (2002), or by numerical simulation, e.g. via the accelerated Stokesian dynamics method (Sierou & Brady 2001) as done by Sierou & Brady (2004). Hence, all terms necessary to determine $\langle V_i(t+t')V_j(t)\rangle$, and thus B_{ij}, are available.

The following tables 6.1, 6.2, and 6.3 are taken in slightly modified form from (Lukassen & Oberlack 2014b). Note that $\langle xx\rangle$ and $\langle \triangle x \triangle x\rangle$ are not listed as this behavior differs from y- and z-direction in case of shear flow in x-direction.

Table 6.1: Summary of sections 2.3.3, 5 and 6 for Brownian particles on τ_p with $\tau_p \ll \tau_D \ll \tau_f$

	on τ_p	
t, $\triangle t$	$< \tau_p$	$> \tau_p$
Brownian velocity	correlated	uncorrelated
configuration		
Diffusion	velocity diffusive	
$\langle yy\rangle$, $\langle zz\rangle$	$\sim t^2$	
$\langle \triangle y \triangle y\rangle$, $\langle \triangle z \triangle z\rangle$	$\sim (\triangle t)^2$	
$\langle U_i U_j\rangle$ with i, j= x, y, z	$\sim t$	constant

Table 6.2: Summary of sections 2.3.3, 5 and 6 for Brownian particles on τ_D with $\tau_p \ll \tau_D \ll \tau_f$

	on τ_D	
t, $\triangle t$	$< \tau_D$	$> \tau_D$
Brownian velocity		
configuration	constant	changing
Diffusion	position short-time	position long-time
$\langle yy \rangle$, $\langle zz \rangle$	$\sim t$	$\sim t$
$\langle \triangle y \triangle y \rangle$, $\langle \triangle z \triangle z \rangle$	$\sim \triangle t$	$\sim \triangle t$
$\langle U_i U_j \rangle$ with i, j= x, y, z	equilibrium	

Table 6.3: Summary of sections 2.3.3, 5 and 6 for non-Brownian particles on τ_f with $\tau_p \ll \tau_f \ll \tau_D$

	on τ_f	
t, $\triangle t$	$< \tau_f$	$> \tau_f$
velocity	correlated	uncorrelated
configuration	constant	changing
Diffusion	colored-noise velocity diffusive	position long-time
$\langle yy \rangle$, $\langle zz \rangle$	$\sim t^2$	$\sim t$
$\langle \triangle y \triangle y \rangle$, $\langle \triangle z \triangle z \rangle$	$\sim (\triangle t)^2$	$\sim \triangle t$
$\langle V_i V_j \rangle$ with i, j= x, y, z	$\sim t$	constant

6.3 The new Fokker-Planck equation in coupled position and velocity space

Inserting the terms into equation 4.43 under the assumption of a composite Markov process, yields:

$$\begin{aligned}
&\frac{\partial \mathcal{P}(x,y,z,V_x,V_y,V_z,t)}{\partial t} = \\
&- \frac{\partial}{\partial x}\Big((U^\infty + V_x)\mathcal{P}\Big) - \frac{\partial}{\partial y}(V_y\mathcal{P}) - \frac{\partial}{\partial z}(V_z\mathcal{P}) \\
&+ \frac{1}{\tau_f}\frac{\partial}{\partial V_x}(V_x\mathcal{P}) + \frac{1}{\tau_f}\frac{\partial}{\partial V_y}(V_y\mathcal{P}) + \frac{1}{\tau_f}\frac{\partial}{\partial V_z}(V_z\mathcal{P}) \\
&+ \frac{1}{2}B_{xx}\frac{\partial^2\mathcal{P}}{\partial V_x \partial V_x} + \frac{1}{2}B_{yy}\frac{\partial^2\mathcal{P}}{\partial V_y \partial V_y} + \frac{1}{2}B_{zz}\frac{\partial^2\mathcal{P}}{\partial V_z \partial V_z} + B_{xy}\frac{\partial^2\mathcal{P}}{\partial V_x \partial V_y}.
\end{aligned} \tag{6.50}$$

The separated behavior in position terms and colored-noise velocity terms due to the composite Markov process is visible. The 3rd and 4th line of equation 6.50 correspond to the Fokker-Planck equation of the Ornstein-Uhlenbeck process of $\boldsymbol{V}$, see also Fokker-Planck equation for an Ornstein-Uhlenbeck process in section 4.2.3.2. As a result, equation 6.50 is a Fokker-Planck equation describing the shear-induced self-diffusion of non-Brownian particles taking into account long correlation times. The analysis

of the different time scale phenomena gives rise to the assumption that in order to fulfill the Markov property a coupled variable of position and colored-noise velocity is necessary.

For clearness, the correlation time of the colored noise is in the following denoted as τ_c. Note that for shear-induced diffusion τ_c is τ_f:

$$\begin{aligned}
&\frac{\partial \mathcal{P}(x,y,z,V_x,V_y,V_z,t)}{\partial t} = \\
&-\frac{\partial}{\partial x}\Big((U^\infty + V_x)\mathcal{P}\Big) - \frac{\partial}{\partial y}(V_y\mathcal{P}) - \frac{\partial}{\partial z}(V_z\mathcal{P}) \\
&+\frac{1}{\tau_c}\frac{\partial}{\partial V_x}(V_x\mathcal{P}) + \frac{1}{\tau_c}\frac{\partial}{\partial V_y}(V_y\mathcal{P}) + \frac{1}{\tau_c}\frac{\partial}{\partial V_z}(V_z\mathcal{P}) \\
&+\frac{1}{2}B_{xx}\frac{\partial^2\mathcal{P}}{\partial V_x \partial V_x} + \frac{1}{2}B_{yy}\frac{\partial^2\mathcal{P}}{\partial V_y \partial V_y} + \frac{1}{2}B_{zz}\frac{\partial^2\mathcal{P}}{\partial V_z \partial V_z} + B_{xy}\frac{\partial^2\mathcal{P}}{\partial V_x \partial V_y}.
\end{aligned} \tag{6.51}$$

In the following, the coupled $U^\infty V_x$-term in equation 6.46, and the respective term in D_{xy}, will be neglected for simplicity which also conforms with the composite Markov process assumption that the behavior in position and velocity is treated separately. In the following, the relation 6.45 for y, z is also used for the xx- and xy-term:

$$B_{ij} = \frac{2D_{ij}}{\tau_c^2} \quad \text{for } i,j = x,y,z. \tag{6.52}$$

6.3.1 Gaussian solution of the colored-noise Fokker-Planck equation

In order to find the probability density $\mathcal{P}(x,y,z,V_x,V_y,V_z,t)$ corresponding to the colored-noise Fokker-Planck equation 6.51 (respectively equation 6.50 as for shear-induced diffusion $\tau_c = \tau_f$), the Gaussian approach given in section 4.2.3.1 according to (van Kampen 2007) is used. This yields the exact probability density, denoted as P_1 in the following. In the authors's publication (Lukassen & Oberlack 2014b), the Gaussian solution has also been derived. This section is based on the corresponding section in the publication, however it is expanded in more detail here. The Gaussian approach from section 4.2.3.1 has also been used by Breedveld et al. (1998) to solve their position-space Fokker-Planck equation. After deriving the Gaussian solution for $\mathcal{P}(x,y,z,V_x,V_y,V_z,t)$ in the present work, this will be compared to the position-space $P(x,y,z,t)$ from (Breedveld et al. 1998). A Gaussian solution analog to the solution from Breedveld et al. (1998) has been derived by Sierou & Brady (2004).
The $\boldsymbol{A}$-matrix used for the Gaussian approach in equation 4.46 yields the following for the present case:

$$\boldsymbol{A} = \begin{bmatrix}
0 & \dot{\gamma} & 0 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 \\
0 & 0 & 0 & \frac{1}{\tau_c} & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{1}{\tau_c} & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{1}{\tau_c}
\end{bmatrix}. \tag{6.53}$$

The appendant $\boldsymbol{B}$-matrix reads:

$$\boldsymbol{B} = \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & B_{xx} & B_{xy} & 0 \\ 0 & 0 & 0 & B_{xy} & B_{yy} & 0 \\ 0 & 0 & 0 & 0 & 0 & B_{zz} \end{bmatrix}. \tag{6.54}$$

As initial condition for $t = 0$, the initial values $x(0), y(0), z(0), V_x(0), V_y(0), V_z(0)$ are set to zero. According to equation 4.49, then, the averaged term in the exponent of the Gaussian solution (equation 4.47) is also zero. Furthermore, a $\boldsymbol{\Xi}$-matrix is derived in correspondence to equation 4.48. This $\boldsymbol{\Xi}$ can be inserted into the Gaussian equation directly, see equation 4.47. The present calculations have been performed with Maple™18 (©Maplesoft, Waterloo Maple Inc.). The resulting term for $\mathcal{P}(x, y, z, V_x, V_y, V_z, t)$ is very extensive and not manageable for an analysis of the appearing terms. For a better handling, an approximation is made in the following. The exponential terms in $\boldsymbol{\Xi}$ (equation 4.48) are neglected when summed with other terms. As an example, the procedure is shown exemplary for the Ξ_{22} component. After integration, the component 6.55 is approximated through 6.56:

$$\Xi_{22} = \frac{1}{2} B_{yy} \tau_c^3 \left(\frac{2t}{\tau_c} - 3 + 4 \exp\left(-\frac{t}{\tau_c} \right) - \exp\left(-\frac{2t}{\tau_c} \right) \right) \tag{6.55}$$

$$\Xi_{22} = \frac{1}{2} B_{yy} \tau_c^3 \left(\frac{2t}{\tau_c} - 3 \right). \tag{6.56}$$

See also equation 3.24 for small times in section 3.1.3 with $U_0 = 0$ (because initial velocities were set to zero above) instead of the equilibrium distribution. For the velocity components, the Ξ_{55} component is regarded as representative here. Equation 6.57 is approximated by equation 6.58:

$$\Xi_{55} = \frac{1}{2} B_{yy} \tau_c \left(1 - \exp\left(-\frac{2t}{\tau_c} \right) \right), \tag{6.57}$$

$$\Xi_{55} = \frac{1}{2} B_{yy} \tau_c. \tag{6.58}$$

Equation 6.57 can also be related to equation 3.16 in section 3.1.3 where $U_0 = 0$ and $t_1 = t_2$. For long times, the regime of equilibrium velocity begins, therefore, the approximated equation 6.58 is related to equation 3.22.

This approximated solution P (i.e. without the exponential terms in $\boldsymbol{\Xi}$) is called P_2 in the following. The index for P here shall indicate the different levels of approximation. Even though the exponential terms in $\boldsymbol{\Xi}$ are neglected, P_2 fulfills the colored-noise Fokker-Planck equation 6.51 with an error of zero. The approximated solution (where the exponential terms were neglected) is also not advantageous to handle so the full P_2 will not be shown here. To give an impression how P_2 looks like, only the leading order terms in t and τ_c are presented below. P_2 is ordered with respect to large times

t. Regarding the equation for short times $t \to 0$, respectively $t < \tau_c$, would not be correct because then the exponential terms that have been neglected above would have considerable influence. The resulting probability density is ordered according to terms of τ_c zeroth order, first order and so on. Whereby here will only be shown zeroth and first order terms. The zeroth order τ_c-terms are organized in the same notation as in the solution of Breedveld et al. (1998) in order to enable the comparison of the two solutions. As will be shown, the zeroth order terms in the present work equal the solution of Breedveld et al. (1998). The solution in (Breedveld et al. 1998), indicated with the index B, reads:

$$P_B(x,y,z,t) = (2\pi)^{-3/2}(a_x a_y a_z)^{-1} \exp\left(-\frac{\left(x - \frac{1}{2}\dot{\gamma}yt - y\frac{D_{xy}}{D_{yy}}\right)^2}{2a_x^2} - \frac{y^2}{2a_y^2} - \frac{z^2}{2a_z^2} \right), \quad (6.59)$$

$$\text{with } a_x^2 = 2D_{xx}t + 2\frac{1}{12}\dot{\gamma}^2 t^3 D_{yy} - 2t\frac{D_{xy}^2}{D_{yy}}, \quad (6.60)$$

$$a_y^2 = 2D_{yy}t, \;\; a_z^2 = 2D_{zz}t, \quad (6.61)$$

whereby here, their solution is modified to starting values in zero (Breedveld et al. have $\triangle$ values). In the following, the solution is given in terms of the position diffusion coefficients D_{ij} in order to compare the equations to the equations from literature. The equations can be written for B_{ij}-terms very easily by just replacing B_{ij} by $2D_{ij}/\tau_c^2$, cf. equation 6.52. P_2 is still too huge to be presented here. Thus, only the leading order terms of P_2 are presented in P_3:

$$\begin{aligned}
P_3(x,y,z,V_x,V_y,V_z,t) = (2\pi)^{-\frac{6}{2}} \Bigg\{ & \left(8\left(D_{xx}t + \frac{1}{12}\dot{\gamma}^2 t^3 D_{yy} - \frac{D_{xy}^2}{D_{yy}}t \right) D_{yy}tD_{zz}t \right) \\
& \left((D_{xx}D_{yy} - D_{xy}^2)D_{zz}\tau_c^{-3} \right) \left(1 - 10\frac{\tau_c}{t} + 40\frac{\tau_c^2}{t^2} + 48\frac{(D_{xx}D_{yy} - D_{xy}^2)\tau_c}{t^3 D_{yy}^2 \dot{\gamma}^2} \right. \\
& \left. + O\left(\frac{\tau_c^2}{t^3}(\tau_c + \frac{\tau_f^2}{t}) \right) \right) \cdot \ldots \Bigg\}^{-1/2} \\
& \exp\Bigg\{ -\frac{1}{2}\left(\frac{(x - \frac{1}{2}\dot{\gamma}yt - y\frac{D_{xy}}{D_{yy}})^2}{a_x^2} + \frac{y^2}{a_y^2} + \frac{z^2}{a_z^2} \right) \\
& - \frac{1}{2}\tau_c \left(\frac{V_x^2}{D_{xx} - D_{xy}^2/D_{yy}} + \frac{V_y^2}{D_{yy} - D_{xy}^2/D_{xx}} + \frac{2V_xV_y}{D_{xy} - D_{xx}D_{yy}/D_{xy}} + \frac{V_z^2}{D_{zz}} \right) \\
& - \frac{1}{2}\tau_c \left(\frac{V_y\dot{\gamma}t(x - \frac{1}{2}y\dot{\gamma}t)}{2\frac{1}{12}\dot{\gamma}^2 t^3 D_{yy}} - \frac{yV_y}{D_{yy}t} - \frac{zV_z}{D_{zz}t} \right) - \ldots \Bigg\}
\end{aligned} \quad (6.62)$$

The (2π) prefactor here is with an exponent $-6/2$ in comparison to Breedveld et al. (1998) because here, the dimension is 6. In arranging the terms with respect to large time t according to equation 6.62, it has to be taken into account that x scales as $t^{3/2}$ and y and z scale as $t^{1/2}$, cf. equations 2.28-2.31 from (Sierou & Brady 2004), (Elrick 1962). The first part of the prefactor, i.e. in the first line of equation 6.62, is the prefactor as also given in (Breedveld et al. 1998). The first term in the second line of 6.62 equals to

the prefactor of a process corresponding to a Fokker-Planck equation for an Ornstein-Uhlenbeck process in $\boldsymbol{V}$. The separate prefactors in highest order for position and velocity variables correspond to the composite Markov process. In zeroth τ_c order, the exponent in equation 6.62 (fourth line) equals exactly the exponent from Breedveld et al. in equation 6.59. It is of order t^0 and τ_c^0. The t^0 results from the scaling of x, y, z, see above. The fifth line of equation 6.62 is of order τ_c and t^0, but since in equilibrium $\langle V_i V_j \rangle$ scales as D_{ij}/τ_c, the fifth line is of the same $\tau_c t$-order as the fourth line. The sixth line is of order τ_c but as x scales as $t^{3/2}$ and y and z scale as $t^{1/2}$ the whole line is of order $\sqrt{\tau_c/t}$.

The P_3 as given in equation 6.62 is not used in the following, it is just shown to get an impression about the terms in P_2 and to show the connection to the position-space Gaussian solution in the respective components. For the purpose of completeness, the P_3 produces an error of $\sqrt{\tau_c^3/t^7}$ when inserted into the colored-noise Fokker-Planck equation 6.51. The P_3 itself is of order $\sqrt{\tau_c^3/t^5}$ whereby $\partial P_3/\partial t$ is also of order $\sqrt{\tau_c^3/t^7}$. Other terms in the colored-noise Fokker-Planck equation as $\frac{1}{\tau_c}\frac{\partial}{\partial V_x}V_x P = \frac{1}{\tau_c}V_x\frac{\partial P}{\partial V_x} + \frac{1}{\tau_c}P$ are of order $\sqrt{\tau_c^3/t^5}$.

6.3.1.1 Moments generated from $P(x, y, z, V_x, V_y, V_z, t)$

In the following shall be given moments generated from P_2, i.e. the approximated probability density, with Maple™18. Note that for the following calculations, not P_3 from equation 6.62 has been used. For the generation of the moments, equation 4.30 from (van Kampen 2007) is used. For the odd moments applies $\langle x \rangle = \int_{-\infty}^{\infty} ... \int_{-\infty}^{\infty} x P_2 dx ... dV_z = 0$ (for the integration over $y = -\infty..\infty$), the same applies for $\langle xxx \rangle$ and so on. When regarding the second moment with respect to τ_c-orders yields:

$$\langle xx \rangle = 2D_{xx}t + 2D_{xy}\dot{\gamma}t^2 + \frac{2}{3}D_{yy}\dot{\gamma}^2 t^3 - \tau_c\left(3D_{xx} + 4D_{xy}\dot{\gamma}t + 2D_{yy}\dot{\gamma}^2 t^2\right) + O(\tau_c^2). \quad (6.63)$$

Throughout the calculations here, it is not allowed to insert $t \to 0$ into the moment equations because the present moments have been calculated via P_2 where the exponential terms are neglected. For $t \to 0$, the exponential terms would have significant influence. Note that the zeroth τ_c order in equation 6.63 is exactly the same as given in (Sierou & Brady 2004) (following (Elrick 1962)), see equation 2.30. For the fourth moment in x-direction applies:

$$\begin{aligned}
\langle x^4 \rangle &= \frac{1}{3}(6D_{xx}t + 6D_{xy}\dot{\gamma}t^2 + 2D_{yy}\dot{\gamma}^2 t^3)^2 + && (6.64)\\
&\quad \tau_c(-36tD_{xx}^2 - 84t^2 D_{xx}D_{xy}\dot{\gamma} - 48t^3 D_{xy}^2\dot{\gamma}^2 - 36t^3 D_{xx}D_{yy}\dot{\gamma}^2 && (6.65)\\
&\quad - 40t^4 D_{xy}D_{yy}\dot{\gamma}^3 - 8t^5 D_{yy}^2\dot{\gamma}^4) + O(\tau_c^2) && (6.66)\\
&= 3\langle xx \rangle^2 + O(\tau_c^2). && (6.67)
\end{aligned}$$

Due to the Gaussian solution, the fourth order cumulants are zero, and the fourth moment can be gained by the second moment, see equation 4.24d. y- and z-direction work analogously. For the colored-noise velocity moment in x-direction applies:

$$\langle V_x^2 \rangle = \frac{D_{xx}}{\tau_c}, \tag{6.68}$$

which is the equilibrium value.

6.4 Short summary of chapter 6

An analysis of the Markov property based on the multiple time scale analysis from chapter 5 shows that a coupled position colored-noise velocity variable is a Markov variable. A model for the colored-noise velocity is introduced based on an Ornstein-Uhlenbeck process. The Fokker-Planck equation in the coupled position colored-noise velocity variable is derived. Additionally, a Gaussian solution is presented and compared to the position-space probability density used so far from Breedveld et al. (1998). In the following chapter 7, the coupled Fokker-Planck equation is reduced to a new position-space formulation and compared to the traditional position-space Fokker-Planck equations for non-Brownian particles in shear flow.

7 The new modified reduced Fokker-Planck equation in position space

In chapter 6, the Fokker-Planck equation in position and colored-noise velocity space was derived with the associated two coupled equations of motion. The probability density $\mathcal{P}(x, y, z, V_x, V_y, V_z, t)$ can be integrated over velocity space to yield the marginal distribution (see equation 4.14, (van Kampen 2007)). An arising question addresses the problem of integrating the whole colored-noise Fokker-Planck equation over velocity space. A Fokker-Planck equation in only the three dimensions of position space of course simplifies the description of the regarded problem. The aim of this chapter is to derive a relation to a new position-space Fokker-Planck equation. Starting from the new colored-noise Fokker-Planck equation 6.50, here will be derived a position-space Fokker-Planck equation which will be compared to the traditional position-space Fokker-Planck equations given in section 6.1.1 which may not be sufficient in the context of non-separated time scales. Throughout the reduction in this chapter, the traditional Fokker-Planck equations in position space used so far can be recovered with additional correction terms. Such an equation as presented in this chapter has not been used to describe the shear-induced diffusion of non-Brownian particles so far. First results on this equation have been presented by the author (Lukassen & Oberlack 2014a). Due to the correction terms, the new position-space Fokker-Planck equation is not a classical Fokker-Planck equation. It is a Fokker-Planck-like type of equation. In the present work, it is simply called Fokker-Planck equation.

7.1 Approach used in the present work

In section 4.3, the reduction of position-velocity space Fokker-Planck equations to pure position-space equations for Brownian particles was given whereby the underlying assumption comprises strongly separated time scales, see (Ermak & McCammon 1978). Since in the non-Brownian case the time scales are not separated, the reduction to position space is not as straightforward. Just integrating the whole colored-noise Fokker-Planck equation over velocity space is not manageable. Consequently, a more structural approach is chosen. In the present work, the approach of Wilemski (1976) is used. Wilemski derived a reduction scheme for the Kramers equation for Brownian particles. The Kramers equation (see (van Kampen 2007)) is the position-velocity Fokker-Planck equation which incorporates an external position-dependent force field, see equation 3.27 in section 3.1.3, and equation 4.67 in section 4.3.2. The approach of Wilemski produces coordinate space equations for higher velocity moments derived from the Kramers equation. Wilemski (1976) solved the moment equations and combined them such that the resulting position-space probability density differential

equation is given in an expansion in inverse powers of the friction. The leading terms in this expansion yield the Smoluchowski equation. According to Wilemski, the hierarchy is not closed but when expanding it in order of the inverse friction coefficient and going up to level five in the moment equations hierarchy results in an exact first order inverse friction coefficient correction. Wilemski states that there have been early attempts to find corrections of friction orders for the Smoluchowski equation, all of which incorporate additional assumptions, cf. sources in (Wilemski 1976), whereby he announces his approach to be free of assumptions. In (Risken 1989) an alternative approach according to Brinkman (1956) and Sack (1956) is presented. Brinkman derived a hierarchy of Laplace transformed equations similar to Wilemski's approach. This is occasionally referred to as Brinkman hierarchy (Risken 1989). This approach is very similar to the approach by Sack. The series of Brinkman (1956) is broken up after the n-th equation, all terms higher than the n-th are neglected, whereby only the thermal equilibrium second order velocity component enters the equation. The result is that the derivation of the first order correction for the Smoluchowski equation, which neglects all moments of second order and higher, only contains the squared thermal velocity (Risken 1989). The more important difference to the Wilemski approach is that the first order moment is eliminated out of the two equations by taking the second time derivative of the zeroth moment equation (Risken 1989). The result is an equation of the form of the telegraph equation (Risken 1989). Due to Sack, the second order time derivative ensures continuity in the first order time derivative when the velocity is rapidly changed. Or in other words, the second order time derivative ensures that a change in the particle density is not instantaneously present (Risken 1989).

In the following, in a first step the approach by Wilemski (1976) will be used to build successive moment equations from the colored-noise Fokker-Planck equation 6.50. In analogy to Wilemski's approach with the series expansion in the inverse friction coefficient, the present expansion is done in the autocorrelation time τ_c of the colored-noise velocity. The prerequisite for the expansion series in terms of τ_c is that the velocity autocorrelation time scale τ_c is considerably smaller than τ_f in order to establish a reasonable expansion. It will be shown that, in accordance with Wilemski, the moment hierarchy up to fifth order is enough to receive a first order correction in the resulting new position-space Fokker-Planck equation. The derivation of the five moment equations yields the zeroth moment and first moment equation. Wilemski combines all higher order moments into a first order in time position-space equation. However, in the present work, the combination of all higher order terms into the zeroth order equation does not lead to correction terms. Due to that, the present work switches to the approach of Brinkman and Sack as shown in (Risken 1989) where the first order moment is eliminated which yields a second order in time equation.
The zeroth order τ_c-terms are exactly the traditional position-space Fokker-Planck equation used so far ((Breedveld et al. 1998), (Sierou & Brady 2004) with constant diffusion coefficients). But of course, it is only possible to neglect the τ_c corrections when τ_c is considerably smaller than the other time scales which in the case of non-separated time scales as in shear-induced diffusion is not the case.

7.1.1 Literature overview on the reduction of coupled equations to position-space equations

Besides the above mentioned approaches, there have been a lot of attempts to reduce coupled position-velocity space Fokker-Planck equations to pure position Fokker-Planck equations. Similar to the works of Brinkman and Sack is also Davies' (Davies 1954), (Davies 1957) approach where for $t \gg \tau_p$ (τ_p is the Brownian velocity relaxation time) the equilibrium value of the velocity autocorrelation is the only second order velocity information. Davies also eliminates the first moment and receives a second order in time equation. The telegraph equation without external forces is solved exactly by Hemmer (1961). Titulaer (1978) derives corrections to the Smoluchowksi equation in an expansion in the inverse friction coefficient by a so-called Chapman-Enskog type approach and receives the same first order correction as Wilemski (1976). A similar approach has also been performed by Wycoff & Balazs (1987), especially with regard to incorporating initial values and calculating the reduced initial values for the resulting Smoluchowski equation. A detailed review on the telegraph equation in the context of deriving the Smoluchowski equation can be found in (Olivares-Robles & García-Colín 1996).

Another way to eliminate the velocity from the Kramers equation by integration results in a memory kernel which describes a non-Markovian process then (Risken 1989). San Miguel & Sancho (1980) work with Brownian particles in colored noise with a position-dependent change in velocity (Brownian harmonic oscillator) and start with the Langevin equation by projecting the phase space Langevin equation to pure position space which yields a non-Markovian Langevin equation. Under certain conditions a corresponding position Fokker-Planck equation can be derived from the non-Markovian Langevin equation. For linear colored-noise Langevin equations, San Miguel & Sancho (1980) derive a Fokker-Planck equation in position space with a time dependent diffusion coefficient which depends on the time integral of the correlation of the colored noise and time integrals of the drift terms, see also (Sancho, San Miguel, Katz & Gunton 1982). This has already been shortly mentioned in the literature overview section 6.1.1. Similar approaches are presented by Hänggi (1978) and Fox (1986). Another possibility to derive friction corrections to the Smoluchowski equation is also presented in (Risken 1989), namely the method of matrix continued fractions. Risken also applies the approach of matrix continued fractions to colored-noise problems and receives similar relations as San Miguel & Sancho (1980). Specific information can also be found in (van Kampen 1989). However, this approach is not followed in the present work. The specific conditions under which such a reduction with integral expressions in the resulting Fokker-Planck equation is possible for the case of the present work have to be investigated. But it appears as a promising procedure to review in the context of reducing the new colored-noise Fokker-Planck equation from chapter 6 in future work, see section 8.2.1.1. There may also be found many other approaches which are not further specified here.

7.2 Precondition on the equation of motion

In the following, it is explicitly assumed that the correlation time of the colored-noise velocity is shorter than the time scale τ_f, i.e. $\tau_c \ll \tau_f$. The regarded time scale of interest is τ_f, which means that the time t is on τ_f, i.e. $t \gg \tau_c$. This enables an expansion in terms of τ_c in relation to the regarded time scale τ_f. As before, it is assumed that the diffusion process with linear in time mean square displacements still starts on τ_f and not on τ_c. The only difference to the situation described before is that the correlation time for the colored noise is assumed as shorter, whereby shorter does not mean infinitely short, i.e. no white noise. The equations of motion corresponding to the problem of a $\tau_c \ll \tau_f$ are used from section 6.2, i.e. equations 6.12-6.13, and from section 6.3, i.e. the colored-noise Fokker-Planck equation 6.51 written in terms of τ_c and the relation between B_{ij} and D_{ij} in equation 6.52.

Scaling of D and B

The scaling of D_{ij} and B_{ij} on τ_c and τ_f has to be derived. The situation described here is similar to the Brownian case. For Brownian particles, the time derivative of mean square displacements on τ_p scales as l^2/τ_p (cf. equations 3.24, 5.4) whereas on τ_D it scales as a^2/τ_D (cf. equation 3.25), cf. e.g. (Dhont 1996). It can be shown that $l^2/\tau_p = a^2/\tau_D$. Transferred to the present case, this means that the D_{ij}, which originally describes the mean square displacements for the system with only one time scale τ_f, can be scaled as:

$$D_{ij} \sim l_1^2/\tau_c = a^2/\tau_f, \tag{7.1}$$

with l_1 as an artificial length scale on τ_c for the present case. For the B_{ij} follows that B_{ij} on τ_c scales as l_1^2/τ_c^3 and on τ_f as $a^2/(\tau_c^2\tau_f)$, cf. equation 6.52.

7.3 The integrated probability density function

In the previous section is derived a Gaussian solution for the colored-noise Fokker-Planck equation, where the exact distribution was denoted as P_1. After neglecting exponential terms of order $\exp(-t/\tau_c)$ in sums, it was called P_2. Integrating P_1 is very extensive. Consequently, the P_2 will be used in the following. Note that P_2 also has not been shown completely. P_2 has only been shown in approximated form denoted as P_3 in equation 6.62 where only the leading order terms have been presented. The probability density $P_2(x, y, z, V_x, V_y, V_z, t)$ can be integrated over the colored-noise velocity space to get a probability density in position space:

$$P^* = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} P_2 dV_x dV_y dV_z. \tag{7.2}$$

In section 4.1.3, the derivation of the $\boldsymbol{\Xi}$ matrix had the precondition that $\boldsymbol{\Xi}$ is positive definite (Risken 1989). Also for the reduced $\boldsymbol{B}$ matrix the same condition applies, since

the colored-noise velocity can be regarded in a separate Fokker-Planck equation. That means for the present case, also the $\boldsymbol{B}$ matrix must be positive definite. Consequently, the following conditions apply: B_{xx}, B_{yy}, B_{zz} are positive and $B_{xx}B_{yy} > B_{xy}^2$. Further, it has to be considered that since the exponential terms are neglected, the P_2 is only valid for large times $t \gg \tau_c$. The integrated probability distribution $P^*(x, y, z, t)$ (equation 7.2) will be compared to the probability distribution of Breedveld et al. (1998) as was given in equation 6.59 in subsection 6.3.1, which was modified by assuming that the starting time is $t = 0$ and that the starting positions are zero for x, y, z. P^* will not be shown completely, but only in the leading order terms denoted as P_h^*. This shall just give an impression about the terms in P^*, in the following, the full P^* will be used. For the calculations, Maple™18 (©Maplesoft, Waterloo Maple Inc.) has been used. For the prefactor, the τ_c^0- and τ_c^1-order terms are presented completely. In the exponent, the τ_c^0-order is shown completely, from the τ_c^1 order only the leading order term in time t is presented. Here, it has to be considered that x grows as $\sqrt{t^3}$ whereas y and z scale as $\sqrt{t}$, cf. section 6.3.1. The integrated probability distribution P_2, i.e. equation 7.2, reads in the leading order terms:

$$
\begin{aligned}
P_h^*(x,y,z,t) = (2\pi)^{-3/2} \Bigg\{ & \left(2D_{xx}t + \frac{2}{12}\dot{\gamma}^2 t^3 D_{yy} - 2\frac{D_{xy}^2}{D_{yy}}t \right) 2D_{yy}t 2D_{zz}t \\
& -\tau_c \Big(5D_{yy}^2\dot{\gamma}^2 t^4 + 36t^2 \left(D_{xx}D_{yy} - D_{xy}^2 \right) D_{zz} \Big) \Bigg\}^{-\frac{1}{2}} \\
& \exp \Bigg\{ -\frac{1}{2} \left(\frac{(x - \frac{1}{2}\dot{\gamma}yt - y\frac{D_{xy}}{D_{yy}})^2}{2D_{xx}t + \frac{2}{12}D_{yy}t^3\dot{\gamma}^2 - 2t\frac{D_{xy}^2}{D_{yy}}} + \frac{y^2}{2D_{yy}t} + \frac{z^2}{2D_{zz}t} \right) \\
& -\tau_c \left(\frac{3}{2}\frac{(3x - \frac{4}{3}y\dot{\gamma}t)^2}{D_{yy}t^4\dot{\gamma}^2} + \frac{1}{3}\frac{y^2}{D_{yy}t^2} + \frac{3}{8}\frac{z^2}{D_{zz}t^2} \right) \Bigg\} .
\end{aligned}
\tag{7.3}
$$

It can be shown, that $P_B(x, y, z, t)$ from Breedveld et al. (1998) (cf. equation 6.59) equals the τ_c^0 order in $P_h^*(x, y, z, t)$. That means that for τ_c tending to zero, this P_h^*, and thus also P^*, corresponds to the traditional probability distribution.

7.4 Model equations

The approach of Wilemski (1976) is used who derives moment equations by multiplying the original Fokker-Planck equation by the velocity in successively higher orders and integrating over velocity space. This enables to investigate the structure of the equation in a more fundamental way than integrating the Fokker-Planck equation numerically. In the present case, the equations look slightly different due to the colored-noise Fokker-Planck equation in contrast to the external force in (Wilemski 1976) but

the approach is the same. In the following, the notation from Wilemski (1976) is used by writing:

$$j_i = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_i P dV_x dV_y dV_z, \tag{7.4}$$

$$P_{ij} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_i V_j P dV_x dV_y dV_z, \tag{7.5}$$

$$Q_{ijk} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_i V_j V_k P dV_x dV_y dV_z, \tag{7.6}$$

$$R_{ijkl} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_i V_j V_k V_l P dV_x dV_y dV_z, \tag{7.7}$$

$$S_{ijklm} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_i V_j V_k V_l V_m P dV_x dV_y dV_z, \tag{7.8}$$

$$T_{ijklmn} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_i V_j V_k V_l V_m V_n P dV_x dV_y dV_z, \tag{7.9}$$

with i, j, k, l, m, n as x, y, z. Note that the P_{ij} with 2 indices i, j from equation 7.5 is not the probability distribution P (which has been used with a single index in the last section). P_{ij} is a matrix and will either be used with 2 indices or bold. Wilemski (1976) states that equations up to fifth order are necessary to get the first order inverse friction correction exactly. As will be shown, this also applies to the present case with τ_c. It can be confirmed that five orders (that means zeroth to fourth order moments) suffice, in this sense the S_{ijklm}-terms and T_{ijklmn}-terms are just listed for completeness, they can be neglected later. The equation of interest is the zeroth moment equation, i.e. the equation for P^*. The successive moment equations are derived in the next section. The resulting equations have a similar form to the equations derived by Wilemski (1976).

For better understanding, it will turn out that the moments scale as follows on τ_f:

$$j_i \sim \frac{a}{\tau_f}, \tag{7.10}$$

$$P_{ij} \sim \frac{a^2}{\tau_c \tau_f}, \tag{7.11}$$

$$Q_{ijk} \sim \frac{a^3}{\tau_c \tau_f^2}, \tag{7.12}$$

$$R_{ijkl} \sim \frac{a^4}{\tau_c^2 \tau_f^2}, \tag{7.13}$$

$$S_{ijklm} \sim \frac{a^5}{\tau_c^2 \tau_f^3}. \tag{7.14}$$

The pre-condition for this scaling is that the sixth order moment, denoted as T_{ijklmn}, does not scale as $\frac{a^6}{\tau_c^4 \tau_f^2}$ but rather as:

$$T_{ijklmn} \sim \frac{a^6}{\tau_c^3 \tau_f^3}. \tag{7.15}$$

7.4.1 Moment equations

The zeroth moment equation for P^* (equation 7.2) results from:

$$\begin{aligned}\frac{\partial}{\partial t}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} P dV_x dV_y dV_z &= -U_x^{\infty}\frac{\partial}{\partial x}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} P dV_x dV_y dV_z \\ -\frac{\partial}{\partial x}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_x P dV_x dV_y dV_z &- \frac{\partial}{\partial y}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_y P dV_x dV_y dV_z \\ -\frac{\partial}{\partial z}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V_z P dV_x dV_y dV_z.\end{aligned} \tag{7.16}$$

It can be shown that the following terms cancel:

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{\partial P}{\partial V_i}dV_i dV_j dV_k = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}[P]_{V_i=-\infty}^{V_i=\infty} dV_j dV_k = 0, \tag{7.17}$$

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{\partial^2 P}{\partial V_i V_j}dV_i dV_j dV_k = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\left[\frac{\partial P}{\partial V_i}\right]_{V_i=-\infty}^{V_i=\infty} dV_j dV_k = 0. \tag{7.18}$$

The $[\,]$-brackets denote the integrated term with the limits given in the superscript and the subscript. In the limits $V_i \to \infty$ and $V_i \to -\infty$, the probability distribution and its derivative approach zero.
The result is the zeroth order equation:

$$\frac{\partial P^*}{\partial t} = -U_x^{\infty}\frac{\partial P^*}{\partial x} - \nabla \cdot \boldsymbol{j}. \tag{7.19}$$

Except for the U_x^{∞}-term, this equation is analog to the zeroth order equation from Wilemski (1976).

The j-equations

For the j_i-terms, the colored-noise Fokker-Planck equation 6.51 is multiplied by V_i and integrated over velocity space. In the following, the integration limits are skipped to keep it short:

$$\begin{aligned}\frac{\partial}{\partial t}\int V_x P dV^3 = -U_x^{\infty}\frac{\partial}{\partial x}\int V_x P dV^3 &- \frac{\partial}{\partial x}\int V_x^2 P dV^3 - \frac{\partial}{\partial y}\int V_x V_y P dV^3 - \frac{\partial}{\partial z}\int V_x V_z P dV^3 \\ &+ \frac{1}{\tau_c}\int V_x\frac{\partial(V_x P)}{\partial V_x}dV^3 + \frac{1}{\tau_c}\int V_x\frac{\partial(V_y P)}{\partial V_y}dV^3 + \frac{1}{\tau_c}\int V_x\frac{\partial(V_z P)}{\partial V_z}dV^3 \\ &+ \frac{1}{2}B_{xx}\int V_x\frac{\partial^2 P}{\partial V_x^2}dV^3 + \frac{1}{2}B_{yy}\int V_x\frac{\partial^2 P}{\partial V_y^2}dV^3 + \frac{1}{2}B_{zz}\int V_x\frac{\partial^2 P}{\partial V_z^2}dV^3 \\ &+ B_{xy}\int V_x\frac{\partial^2 P}{\partial V_x V_y}dV^3.\end{aligned} \tag{7.20}$$

Here has to be considered that:

$$\int\int V_j \int \frac{\partial(V_i P)}{\partial V_i} dV_i dV_j dV_k = \int\int V_j [V_i P]_{-\infty}^{\infty} dV_j dV_k = 0, \tag{7.21}$$

$$\int V_i \frac{\partial(V_i P)}{\partial V_i} dV_i = \int \frac{\partial(V_i^2 P)}{\partial V_i} dV_i - \int P V_i dV_i = 0 - \int P V_i dV_i. \tag{7.22}$$

Both conditions are fulfilled when P for $V_i \to \infty$ approaches zero faster than V_i^2 approaches ∞. This can be confirmed with Maple™18 (©Maplesoft, Waterloo Maple Inc.) by inserting P_2.

$$\frac{\partial j_x}{\partial t} = -U_x^{\infty} \frac{\partial j_x}{\partial x} - \nabla \cdot \boldsymbol{P}_x - \frac{1}{\tau_c} j_x, \tag{7.23}$$

$$\frac{\partial j_y}{\partial t} = -U_x^{\infty} \frac{\partial j_y}{\partial x} - \nabla \cdot \boldsymbol{P}_y - \frac{1}{\tau_c} j_y, \tag{7.24}$$

$$\frac{\partial j_z}{\partial t} = -U_x^{\infty} \frac{\partial j_z}{\partial x} - \nabla \cdot \boldsymbol{P}_z - \frac{1}{\tau_c} j_z. \tag{7.25}$$

The j_i equations are analog to Wilemski's equation of first order except for the U_x^{∞}-term. The difference is that the U_x^{∞}-term enters with a $\partial j_i / \partial x$-term while in (Wilemski 1976) the external force enters just with P^*. This difference will require a transformation for the integration later. The $\boldsymbol{P}$ consists of 9 components, thus $\boldsymbol{P}_x$, $\boldsymbol{P}_y$, $\boldsymbol{P}_z$ consist of 3 components, respectively.

The $\boldsymbol{P}$-equations

The $\boldsymbol{P}$ matrix is symmetric, which means that the following components, P_{x1}, P_{x2}, P_{x3}, P_{y2}, P_{y3}, P_{z3} have to be computed, whereas P_{y1}, P_{z1}, P_{z2} are included. Following the approach from (Wilemski 1976), the colored-noise Fokker-Planck equation is multiplied by $V_i V_j$ and integrated over velocity space. Further, the following rules apply:

$$\int \frac{\partial(V_i P)}{\partial V_i} V_i^2 dV_i = \int \frac{\partial(V_i^3 P)}{\partial V_i} dV_i - \int 2 P V_i^2 dV_i = 0 - \int 2 P V_i^2 dV_i, \tag{7.26}$$

$$\int \frac{\partial^2 P}{\partial V_i^2} V_i^2 dV_i = \int \left(\frac{\partial^2 (P V_i^2)}{\partial V_i^2} - 4 V_i \frac{\partial P}{\partial V_i} - 2P \right) dV_i \tag{7.27}$$

$$= -\int 4 V_i \frac{\partial P}{\partial V_i} dV_i - \int 2 P dV_i = -4 \int \frac{\partial V_i P}{\partial V_i} dV_i + 2 \int P dV_i \tag{7.28}$$

$$= 2 \int P dV_i. \tag{7.29}$$

The term in equation 7.26 can be transformed to $-2P_{ii}$. The term in equation 7.29 yields $2P^*$, when integrated over all velocity components. The resulting moment equations for second order read:

$$\frac{\partial P_{x1}}{\partial t} = -U_x^{\infty} \frac{\partial P_{x1}}{\partial x} - \nabla \cdot \boldsymbol{Q}_{o11} - \frac{2}{\tau_c} P_{x1} + B_{xx} P^*, \tag{7.30}$$

$$\frac{\partial P_{x2}}{\partial t} = -U_x^\infty \frac{\partial P_{x2}}{\partial x} - \nabla \cdot \boldsymbol{Q}_{o12} - \frac{2}{\tau_c} P_{x2} + B_{xy} P^*, \tag{7.31}$$

$$\frac{\partial P_{x3}}{\partial t} = -U_x^\infty \frac{\partial P_{x3}}{\partial x} - \nabla \cdot \boldsymbol{Q}_{o13} - \frac{2}{\tau_c} P_{x3}, \tag{7.32}$$

$$\frac{\partial P_{y2}}{\partial t} = -U_x^\infty \frac{\partial P_{y2}}{\partial x} - \nabla \cdot \boldsymbol{Q}_{o22} - \frac{2}{\tau_c} P_{y2} + B_{yy} P^*, \tag{7.33}$$

$$\frac{\partial P_{y3}}{\partial t} = -U_x^\infty \frac{\partial P_{y3}}{\partial x} - \nabla \cdot \boldsymbol{Q}_{o23} - \frac{2}{\tau_c} P_{y3}, \tag{7.34}$$

$$\frac{\partial P_{z3}}{\partial t} = -U_x^\infty \frac{\partial P_{z3}}{\partial x} - \nabla \cdot \boldsymbol{Q}_{o33} - \frac{2}{\tau_c} P_{z3} + B_{zz} P^*. \tag{7.35}$$

As above, these equations are similar to the second order equations from Wilemski except for the U_x^∞-terms. The index o for the $\boldsymbol{Q}$-terms refers to the gradient, whereas the second and third index of $\boldsymbol{Q}_{oij}$ refer to the particular P_{ij} in the very same equation.

The Q-equations

$\boldsymbol{Q}$ exhibits 27 components, whereby the $\boldsymbol{Q}$-tensor also is symmetric. Due to symmetry, the following components have to be computed: Q_{111}, Q_{112}, Q_{113}, Q_{212}, Q_{213}, Q_{222}, Q_{223}, Q_{331}, Q_{332}, Q_{333}, whereby these include the components Q_{121}, Q_{211}, Q_{131}, Q_{311}, Q_{221}, Q_{122}, Q_{123}, Q_{132}, Q_{312}, Q_{321}, Q_{231}, Q_{322}, Q_{232}, Q_{313}, Q_{133}, Q_{233}, Q_{323}. The third order moment equations read:

$$\frac{\partial Q_{122}}{\partial t} = -U_x^\infty \frac{\partial Q_{122}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o122} - \frac{3}{\tau_c} Q_{122} + B_{yy} j_x + 2 B_{xy} j_y, \tag{7.36}$$

$$\frac{\partial Q_{113}}{\partial t} = -U_x^\infty \frac{\partial Q_{113}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o113} - \frac{3}{\tau_c} Q_{113} + B_{xx} j_z, \tag{7.37}$$

$$\frac{\partial Q_{222}}{\partial t} = -U_x^\infty \frac{\partial Q_{222}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o222} - \frac{3}{\tau_c} Q_{222} + 3 B_{yy} j_y, \tag{7.38}$$

$$\frac{\partial Q_{123}}{\partial t} = -U_x^\infty \frac{\partial Q_{123}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o123} - \frac{3}{\tau_c} Q_{123} + B_{xy} j_z, \tag{7.39}$$

$$\frac{\partial Q_{111}}{\partial t} = -U_x^\infty \frac{\partial Q_{111}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o111} - \frac{3}{\tau_c} Q_{111} + 3 B_{xx} j_x, \tag{7.40}$$

$$\frac{\partial Q_{112}}{\partial t} = -U_x^\infty \frac{\partial Q_{112}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o112} - \frac{3}{\tau_c} Q_{112} + B_{xx} j_y + 2 B_{xy} j_x, \tag{7.41}$$

$$\frac{\partial Q_{233}}{\partial t} = -U_x^\infty \frac{\partial Q_{233}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o233} - \frac{3}{\tau_c} Q_{233} + B_{zz} j_y, \tag{7.42}$$

$$\frac{\partial Q_{333}}{\partial t} = -U_x^\infty \frac{\partial Q_{333}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o333} - \frac{3}{\tau_c} Q_{333} + 3 B_{zz} j_z, \tag{7.43}$$

$$\frac{\partial Q_{133}}{\partial t} = -U_x^\infty \frac{\partial Q_{133}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o133} - \frac{3}{\tau_c} Q_{133} + B_{zz} j_x, \tag{7.44}$$

$$\frac{\partial Q_{223}}{\partial t} = -U_x^\infty \frac{\partial Q_{223}}{\partial x} - \nabla \cdot \boldsymbol{R}_{o223} - \frac{3}{\tau_c} Q_{223} + B_{yy} j_z. \tag{7.45}$$

As pointed out above, the index o of the $\boldsymbol{R}_{oijk}$ refers to the gradient, while the other indices refer to the respective Q_{ijk}.

The $\boldsymbol{R}$-equations

The $\boldsymbol{R}$-equations are 15 independent equations, the other components are included due to symmetry:

$$\frac{\partial R_{1111}}{\partial t} = -U_x^\infty \frac{\partial R_{1111}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1111} - \frac{4}{\tau_c} R_{1111} + 6B_{xx}P_{x1}, \tag{7.46}$$

$$\frac{\partial R_{1112}}{\partial t} = -U_x^\infty \frac{\partial R_{1112}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1112} - \frac{4}{\tau_c} R_{1112} + 3B_{xx}P_{x2} + 3B_{xy}P_{x1}, \tag{7.47}$$

$$\frac{\partial R_{1113}}{\partial t} = -U_x^\infty \frac{\partial R_{1113}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1113} - \frac{4}{\tau_c} R_{1113} + 3B_{xx}P_{x3}, \tag{7.48}$$

$$\frac{\partial R_{1222}}{\partial t} = -U_x^\infty \frac{\partial R_{1222}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1222} - \frac{4}{\tau_c} R_{1222} + 3B_{xy}P_{y2} + 3B_{yy}P_{x2}, \tag{7.49}$$

$$\frac{\partial R_{1122}}{\partial t} = -U_x^\infty \frac{\partial R_{1122}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1122} - \frac{4}{\tau_c} R_{1122} + B_{xx}P_{y2} + B_{yy}P_{x1} + 4B_{xy}P_{x2}, \tag{7.50}$$

$$\frac{\partial R_{1133}}{\partial t} = -U_x^\infty \frac{\partial R_{1133}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1133} - \frac{4}{\tau_c} R_{1133} + B_{xx}P_{z3} + B_{zz}P_{x1}, \tag{7.51}$$

$$\frac{\partial R_{1123}}{\partial t} = -U_x^\infty \frac{\partial R_{1123}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1123} - \frac{4}{\tau_c} R_{1123} + B_{xx}P_{y3} + 2B_{xy}P_{x3}, \tag{7.52}$$

$$\frac{\partial R_{1223}}{\partial t} = -U_x^\infty \frac{\partial R_{1223}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1223} - \frac{4}{\tau_c} R_{1223} + 2B_{xy}P_{y3} + B_{yy}P_{x3}, \tag{7.53}$$

$$\frac{\partial R_{1233}}{\partial t} = -U_x^\infty \frac{\partial R_{1233}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1233} - \frac{4}{\tau_c} R_{1233} + B_{xy}P_{z3} + B_{zz}P_{x2}, \tag{7.54}$$

$$\frac{\partial R_{2222}}{\partial t} = -U_x^\infty \frac{\partial R_{2222}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o2222} - \frac{4}{\tau_c} R_{2222} + 6B_{yy}P_{y2}, \tag{7.55}$$

$$\frac{\partial R_{3333}}{\partial t} = -U_x^\infty \frac{\partial R_{3333}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o3333} - \frac{4}{\tau_c} R_{3333} + 6B_{zz}P_{z3}, \tag{7.56}$$

$$\frac{\partial R_{1333}}{\partial t} = -U_x^\infty \frac{\partial R_{1333}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o1333} - \frac{4}{\tau_c} R_{1333} + 3B_{zz}P_{x3}, \tag{7.57}$$

$$\frac{\partial R_{2233}}{\partial t} = -U_x^\infty \frac{\partial R_{2233}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o2233} - \frac{4}{\tau_c} R_{2233} + B_{yy}P_{z3} + B_{zz}P_{y2}, \tag{7.58}$$

$$\frac{\partial R_{2223}}{\partial t} = -U_x^\infty \frac{\partial R_{2223}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o2223} - \frac{4}{\tau_c} R_{2223} + 3B_{yy}P_{y3}, \tag{7.59}$$

$$\frac{\partial R_{2333}}{\partial t} = -U_x^\infty \frac{\partial R_{2333}}{\partial x} - \nabla \cdot \boldsymbol{S}_{o2333} - \frac{4}{\tau_c} R_{2333} + 3B_{zz}P_{y3}. \tag{7.60}$$

For the moment equations for $\boldsymbol{S}$, here will only be shown an example. They are not necessary for the first order correction with respect to τ_c.

$$\frac{\partial S_{11222}}{\partial t} = -U_x^\infty \frac{\partial S_{11222}}{\partial x} - \nabla \cdot \boldsymbol{T}_{o11222} - \frac{5}{\tau_c} S_{11222} + B_{xx}Q_{222} + \ldots, \tag{7.61}$$

The $\boldsymbol{T}_{o11222}$ is a placeholder for the next higher moment.

7.4.2 Solving the differential moment equations

In the second step, the moment equations are solved. As the moment equations in the present work look slightly different from the equations for a Kramers equation (Wilemski 1976), here, a transformation of variables has to be used. For the method of characteristics, the variables are transformed as follows:

$$t_* = t,\ x_* = x - U_x^\infty t,\ y_* = y,\ z_* = z, \tag{7.62}$$

$$\frac{\partial}{\partial t} = \frac{\partial t_*}{\partial t}\frac{\partial}{\partial t_*} + \frac{\partial x_*}{\partial t}\frac{\partial}{\partial x_*} + \frac{\partial y_*}{\partial t}\frac{\partial}{\partial y_*} + \frac{\partial z_*}{\partial t}\frac{\partial}{\partial z_*} = \frac{\partial}{\partial t_*} - U_x^\infty \frac{\partial}{\partial x_*}, \tag{7.63}$$

$$\frac{\partial}{\partial x} = \frac{\partial}{\partial x_*}, \tag{7.64}$$

$$\frac{\partial}{\partial y} = \frac{\partial t_*}{\partial y}\frac{\partial}{\partial t_*} + \frac{\partial x_*}{\partial y}\frac{\partial}{\partial x_*} + \frac{\partial y_*}{\partial y}\frac{\partial}{\partial y_*} + \frac{\partial z_*}{\partial y}\frac{\partial}{\partial z_*} = -\dot{\gamma}t\frac{\partial}{\partial x_*} + \frac{\partial}{\partial y_*}, \tag{7.65}$$

$$\frac{\partial}{\partial z} = \frac{\partial}{\partial z_*}, \tag{7.66}$$

with $U_x^\infty = \dot{\gamma}y$. Applying this transformation to the differential equations results in equations without the derivation with respect to x. The $*$ indicates the transformed variables.

The j-equations

Transforming j_x according to equations 7.62- 7.66, yields:

$$\frac{\partial j_x}{\partial t_*} - U_x^\infty \frac{\partial j_x}{\partial x_*} = -U_x^\infty \frac{\partial j_x}{\partial x_*} - \frac{\partial}{\partial x_*}P_{x1} - \frac{\partial}{\partial y_*}P_{x2} + \dot{\gamma}t\frac{\partial}{\partial x_*}P_{x2} - \frac{\partial}{\partial z_*}P_{x3} - \frac{1}{\tau_c}j_x, \tag{7.67}$$

$$\frac{\partial j_x}{\partial t_*} = -\nabla_* \cdot \boldsymbol{P}_x + \frac{t}{\tau_f}\frac{\partial}{\partial x_*}P_{x2} - \frac{1}{\tau_c}j_x. \tag{7.68}$$

After the transformation, this equation has a form similar to the equations in (Wilemski 1976). So here, the same way of integrating this equation is used following Wilemski (integration by parts):

$$j_x = -\int_0^{t_*} \exp\left(\frac{-(t_* - \tau)}{\tau_c}\right)\left(\nabla_* \cdot \boldsymbol{P}_x - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}P_{x2}\right)d\tau \tag{7.69}$$

$$= i.t. - \left[\tau_c\left(\nabla_* \cdot \boldsymbol{P}_x - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}P_{x2}\right) - \int_0^{t_*} \tau_c \exp\left(\frac{-(t_* - \tau)}{\tau_c}\right)\frac{\partial}{\partial \tau}\left(\nabla_* \cdot \boldsymbol{P}_x - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}P_{x2}\right)d\tau\right] \tag{7.70}$$

$$= i.t. - \left[\tau_c\left(\nabla_* \cdot \boldsymbol{P}_x - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}P_{x2}\right) - \tau_c^2\frac{\partial}{\partial t_*}\left(\nabla_* \cdot \boldsymbol{P}_x - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}P_{x2}\right)\right.$$
$$\left. + \int_0^{t_*} \tau_c^2 \exp\left(\frac{-(t_* - \tau)}{\tau_c}\right)\frac{\partial^2}{\partial \tau^2}\left(\nabla_* \cdot \boldsymbol{P}_x - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}P_{x2}\right)d\tau\right]. \tag{7.71}$$

i.t. are, in compliance with Wilemski (1976), the initial terms, i.e. at time $t = 0$. According to Wilemski, the initial terms enter with an exponential term, transferred to the present case this means $\exp(-t/\tau_c)$. As here shall be regarded times $t \gg \tau_c$, the initial terms are not further treated in the following, so they will not be mentioned anymore. Furthermore, in the present case, the initial position and velocity variables were all set to zero in section 6.3.1.
After re-substituting the highest order terms result in:

$$j_x = -\tau_c \nabla \cdot \boldsymbol{P}_x + \tau_c^2 \left(\frac{\partial}{\partial t} \nabla \cdot \boldsymbol{P}_x + U_x^\infty \frac{\partial}{\partial x} \nabla \cdot \boldsymbol{P}_x \right). \tag{7.72}$$

The τ_c^2-terms in the brackets scale as $1/\tau_f$ because the regarded time t is on the time scale τ_f, thus the time derivative $\partial/\partial t$ scales as $1/\tau_f$, and the shear velocity U_x^∞ also scales as $1/\tau_f$. Consequently, the first order term of j_x reads:

$$j_x = -\tau_c \nabla \cdot \boldsymbol{P}_x, \tag{7.73}$$

with first order $\boldsymbol{P}_x$. The second order of j_x needs also the second order of $\boldsymbol{P}_x$ as this will be included in the τ_c-term in equation 7.72. The equations for j_y and j_z are analog:

$$j_y = -\tau_c \nabla \cdot \boldsymbol{P}_y + \tau_c^2 \left(\frac{\partial}{\partial t} \nabla \cdot \boldsymbol{P}_y + U_x^\infty \frac{\partial}{\partial x} \nabla \cdot \boldsymbol{P}_y \right), \tag{7.74}$$

$$j_z = -\tau_c \nabla \cdot \boldsymbol{P}_z + \tau_c^2 \left(\frac{\partial}{\partial t} \nabla \cdot \boldsymbol{P}_z + U_x^\infty \frac{\partial}{\partial x} \nabla \cdot \boldsymbol{P}_z \right). \tag{7.75}$$

The $\boldsymbol{P}$-equations

The equations for $\boldsymbol{P}$ are solved the same way. Here, the procedure is shown for the $x1$ component of $\boldsymbol{P}$, i.e. P_{x1}:

$$\frac{\partial P_{x1}}{\partial t_*} - U_x^\infty \frac{\partial P_{x1}}{\partial x_*} = -U_x^\infty \frac{\partial P_{x1}}{\partial x_*} - \nabla_* \cdot \boldsymbol{Q}_{o11} + \frac{t_*}{\tau_f} \frac{\partial}{\partial x_*} Q_{211} - \frac{2}{\tau_c} P_{x1} + B_{xx} P^*, \tag{7.76}$$

$$\frac{\partial P_{x1}}{\partial t_*} = -\nabla_* \cdot \boldsymbol{Q}_{o11} + \frac{t_*}{\tau_f} \frac{\partial}{\partial x_*} Q_{211} - \frac{2}{\tau_c} P_{x1} + B_{xx} P^*. \tag{7.77}$$

Note that the superscript $*$ in P^* has nothing to do with the transformation. The transformation according to equations 7.62-7.66 is indicated through the subscript $*$. Following the integration procedure in (Wilemski 1976), yields:

$$P_{x1} = -\int_0^{t_*} \exp\left(\frac{-2(t_* - \tau)}{\tau_c} \right) \left(\nabla_* \cdot \boldsymbol{Q}_{o11} - \frac{\tau}{\tau_f} \frac{\partial}{\partial x_*} Q_{211} - B_{xx} P^* \right) d\tau \tag{7.78}$$

$$= -\left[\frac{\tau_c}{2} \left(\nabla_* \cdot \boldsymbol{Q}_{o11} - \frac{t_*}{\tau_f} \frac{\partial}{\partial x_*} Q_{211} - B_{xx} P^* \right) - \frac{\tau_c^2}{4} \frac{\partial}{\partial t_*} \left(\nabla_* \cdot \boldsymbol{Q}_{o11} - \frac{t_*}{\tau_f} \frac{\partial}{\partial x_*} Q_{211} - B_{xx} P^* \right) + \ldots \right]. \tag{7.79}$$

More details on the integration can be found in appendix D.1. The terms with the subscript $*$ are re-substituted:

$$P_{x1} = -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o11} - B_{xx}P^*\right) + \frac{\tau_c^2}{4}\left(\frac{\partial}{\partial t}\left(\nabla\cdot\boldsymbol{Q}_{o11} - B_{xx}P^*\right) + U_x^\infty\frac{\partial}{\partial x}\left(\nabla\cdot\boldsymbol{Q}_{o11} - B_{xx}P^*\right)\right) - \ldots \tag{7.80}$$

In order to compute 2 orders of $\boldsymbol{j}$, also $\boldsymbol{P}$ is required to the second order. Note that B_{xx} scales as $2D_{xx}/\tau_c^2$. The first 2 orders of P_{x1} include $O(1/\tau_c)$-terms and $O(1)$-terms. In the second line of equation 7.80, the $B_{xx}P^*$-component dominates over the $\nabla\cdot\boldsymbol{Q}_{o11}$-terms as will be shown below. Consequently, from the second line, only the $B_{xx}P^*$-terms remain.
All $\boldsymbol{P}$ equations read:

$$P_{x1} = -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o11} - B_{xx}P^*\right) - \frac{\tau_c^2}{4}\left(B_{xx}\frac{\partial P^*}{\partial t} + U_x^\infty B_{xx}\frac{\partial P^*}{\partial x}\right), \tag{7.81}$$

$$P_{x2} = -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o12} - B_{xy}P^*\right) - \frac{\tau_c^2}{4}\left(B_{xy}\frac{\partial P^*}{\partial t} + U_x^\infty B_{xy}\frac{\partial P^*}{\partial x}\right), \tag{7.82}$$

$$P_{x3} = -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o13}\right), \tag{7.83}$$

$$P_{y2} = -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o22} - B_{yy}P^*\right) - \frac{\tau_c^2}{4}\left(B_{yy}\frac{\partial P^*}{\partial t} + U_x^\infty B_{yy}\frac{\partial P^*}{\partial x}\right), \tag{7.84}$$

$$P_{y3} = -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o23}\right), \tag{7.85}$$

$$P_{z3} = -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o33} - B_{zz}P^*\right) - \frac{\tau_c^2}{4}\left(B_{zz}\frac{\partial P^*}{\partial t} + U_x^\infty B_{zz}\frac{\partial P^*}{\partial x}\right). \tag{7.86}$$

As will be pointed out below, $\boldsymbol{Q}$ is one order lower than the other terms, so the highest order of $\boldsymbol{P}$ can be derived directly from the equations 7.81-7.86 as:

$$P_{x1}^h = \frac{\tau_c}{2}B_{xx}P^* = \frac{D_{xx}}{\tau_c}P^*, \tag{7.87}$$

$$P_{x2}^h = \frac{\tau_c}{2}B_{xy}P^* = \frac{D_{xy}}{\tau_c}P^*, \tag{7.88}$$

$$P_{x3}^h = 0, \tag{7.89}$$

$$P_{y2}^h = \frac{\tau_c}{2}B_{yy}P^* = \frac{D_{yy}}{\tau_c}P^*, \tag{7.90}$$

$$P_{y3}^h = 0, \tag{7.91}$$

$$P_{z3}^h = \frac{\tau_c}{2}B_{zz}P^* = \frac{D_{zz}}{\tau_c}P^*. \tag{7.92}$$

These equations show that $\boldsymbol{P}$ scales in highest order as $a^2/(\tau_c\tau_f)$ as predicted in equation 7.11. The D_{ij} scales as a^2/τ_f as given in equation 7.1.

The $\boldsymbol{Q}$-equations

In the following, the equations for $\boldsymbol{Q}$ are solved:

$$\frac{\partial Q_{122}}{\partial t_*} - U_x^\infty \frac{\partial Q_{122}}{\partial x_*} = -U_x^\infty \frac{\partial Q_{122}}{\partial x_*} - \nabla_* \cdot \boldsymbol{R}_{o122} + \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - \frac{3}{\tau_c}Q_{122} + B_{yy}j_x + 2B_{xy}j_y, \tag{7.93}$$

$$\frac{\partial Q_{122}}{\partial t_*} = -\nabla_* \cdot \boldsymbol{R}_{o122} + \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - \frac{3}{\tau_c}Q_{122} + B_{yy}j_x + 2B_{xy}j_y. \tag{7.94}$$

Integration yields, cf. (Wilemski 1976):

$$Q_{122} = -\int_0^{t_*} \exp\left(\frac{-3(t_*-\tau)}{\tau_c}\right)\left(\nabla_* \cdot \boldsymbol{R}_{o122} - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)d\tau \tag{7.95}$$

$$\begin{aligned} = -\Big[&\frac{\tau_c}{3}\left(\nabla \cdot \boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right) \\ &- \frac{\tau_c^2}{9}\frac{\partial}{\partial t}\left(\nabla \cdot \boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right) + \frac{\tau_c^2}{9}U_x^\infty\frac{\partial}{\partial x}\left(\nabla \cdot \boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right) \\ &+ \int_0^t \frac{\tau_c^2}{9}\exp\left(\frac{-3(t-\tau)}{\tau_c}\right)\frac{\partial^2}{\partial\tau^2}\left(\nabla \cdot \boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)d\tau\Big]. \end{aligned} \tag{7.96}$$

More details can be found in appendix D.2. In equation 7.96, the τ_c^2-terms can be neglected since Q_{122} shall only be determined in first order τ_c. The time derivative is on the time scale τ_f and thus does not reduce the order of the τ_c^2-terms. Consequently, in lowest τ_c-order, it follows for Q_{122} (after re-substitution of the variables):

$$Q_{122} = \frac{\tau_c}{3}(-\nabla \cdot \boldsymbol{R}_{o122} + B_{yy}j_x + 2B_{xy}j_y). \tag{7.97}$$

The other $\boldsymbol{Q}$-terms are analog but not shown here. They are solved directly in the next section 7.4.3.

7.4.3 Solving the equations iteratively

In the following, the equations are successively solved by inserting the terms for $\boldsymbol{j}, \boldsymbol{P}, \boldsymbol{Q}, \boldsymbol{R}$.

The R equations

For the moment equations in $\boldsymbol{R}$, the $\boldsymbol{S}$ are assumed to be negligible. The $\boldsymbol{S}$ equation (cf. equation 7.61) contains, besides the unknown $\boldsymbol{T}$-term, in leading order $\boldsymbol{S} = \frac{\tau_c}{5}B_{ij}Q_{klm} = \frac{2}{5\tau_c}D_{ij}Q_{klm}$. As will be shown below, when $\boldsymbol{T}$ fulfills equation 7.15, i.e. does not contain terms τ_c^{-k}, with $k > 3$, then $\boldsymbol{Q}$ is of leading order $a^3/(\tau_c\tau_f^2)$. Consequently, $\boldsymbol{S}$ is of order $a^5/(\tau_c^2\tau_f^3)$ as predicted in equation 7.14.
The $\boldsymbol{R}$ equations are solved in the first order. The time derivative in the $\boldsymbol{R}$ equations is

one order higher than the $B_{kl}P_{ij}$.
Since only the highest order $\boldsymbol{R}$ term is requested here, the time derivative can be neglected. Further, the P_{ij} terms are multiplied by $1/\tau_c^2$ through the $\boldsymbol{B}$-terms, thus the last term in the $\boldsymbol{R}$ equations is of order $a^4/(\tau_c^3\tau_f^2)$. Consequently, also the contributions from $\boldsymbol{S}$ are negligible. Solving the first equation for $\boldsymbol{R}$ yields, after inserting the highest order from $\boldsymbol{P}$ (equations 7.87-7.92):

$$R_{1111} = \frac{6}{4}\tau_c B_{xx} P_{x1} = \frac{6}{4}\frac{1}{\tau_c} 2 D_{xx} D_{xx} \frac{1}{\tau_c} P^* = \frac{3}{\tau_c^2} D_{xx} D_{xx} P^*, \tag{7.98}$$

$$R_{1112} = \frac{3}{\tau_c^2} D_{xx} D_{xy} P^*, \tag{7.99}$$

$$R_{1113} = \frac{3}{4}\tau_c B_{xx} P_{x3} = 0, \tag{7.100}$$

$$R_{1222} = \frac{3}{\tau_c^2} D_{xy} D_{yy} P^*, \tag{7.101}$$

$$R_{1122} = \frac{1}{\tau_c^2} D_{xx} D_{yy} P^* + \frac{2}{\tau_c^2} D_{xy} D_{xy} P^*, \tag{7.102}$$

$$R_{1133} = \frac{1}{\tau_c^2} D_{xx} D_{zz} P^*, \tag{7.103}$$

$$R_{1123} = \frac{1}{4}\tau_c B_{xx} P_{y3} + \frac{1}{2}\tau_c B_{xy} P_{x3} = 0, \tag{7.104}$$

$$R_{1223} = \frac{1}{2}\tau_c B_{xy} P_{y3} + \frac{1}{4}\tau_c B_{yy} P_{x3} = 0, \tag{7.105}$$

$$R_{1233} = \frac{1}{\tau_c^2} D_{xy} D_{zz} P^*, \tag{7.106}$$

$$R_{2222} = \frac{3}{\tau_c^2} D_{yy} D_{yy} P^*, \tag{7.107}$$

$$R_{3333} = \frac{3}{\tau_c^2} D_{zz} D_{zz} P^*, \tag{7.108}$$

$$R_{1333} = \frac{3}{4}\tau_c B_{zz} P_{x3} = 0, \tag{7.109}$$

$$R_{2233} = \frac{1}{\tau_c^2} D_{yy} D_{zz} P^*, \tag{7.110}$$

$$R_{2223} = \frac{3}{4}\tau_c B_{yy} P_{y3} = 0, \tag{7.111}$$

$$R_{2333} = \frac{3}{4}\tau_c B_{zz} P_{y3} = 0. \tag{7.112}$$

More details can be found in appendix D.3. The $\boldsymbol{R}$ scale as $a^4/(\tau_c^2\tau_f^2)$ as assumed in equation 7.13.

The Q equations

The $\boldsymbol{Q}$ shall be solved in lowest order. For the $\boldsymbol{Q}$, the lowest order in $\boldsymbol{j}$ is needed (from equations 7.72-7.75). This in turn can be gained via the lowest order in $\boldsymbol{P}$ (from equations 7.87-7.92) and yields:

$$j_x^h = -\tau_c \nabla \cdot \boldsymbol{P}_x = -D_{xx}\frac{\partial P^*}{\partial x} - D_{xy}\frac{\partial P^*}{\partial y}, \tag{7.113}$$

$$j_y^h = -\tau_c \nabla \cdot \boldsymbol{P}_y = -D_{xy}\frac{\partial P^*}{\partial x} - D_{yy}\frac{\partial P^*}{\partial y}, \tag{7.114}$$

$$j_z^h = -\tau_c \nabla \cdot \boldsymbol{P}_z = -D_{zz}\frac{\partial P^*}{\partial z}. \tag{7.115}$$

This shows that $\boldsymbol{j}$ scales as a/τ_f as given in equation 7.10.

Further, the solved $\boldsymbol{R}$ terms are needed from equations 7.98-7.112. Inserting these terms into the equation 7.97 for Q_{122}, yields:

$$\begin{aligned} Q_{122} = \frac{\tau_c}{3}\Bigg(& -\frac{\partial}{\partial x}\left(\frac{1}{\tau_c^2}D_{xx}D_{yy}P^* + \frac{2}{\tau_c^2}D_{xy}D_{xy}P^*\right) \\ & -\frac{\partial}{\partial y}\left(\frac{3}{\tau_c^2}D_{xy}D_{yy}P^*\right) - 0 + B_{yy}j_x + 2B_{xy}j_y\Bigg) \end{aligned} \tag{7.116}$$

$$\begin{aligned} = \frac{\tau_c}{3}\Bigg(& -\frac{1}{\tau_c^2}D_{xx}D_{yy}\frac{\partial P^*}{\partial x} - \frac{2}{\tau_c^2}D_{xy}D_{xy}\frac{\partial P^*}{\partial x} \\ & -\frac{3}{\tau_c^2}D_{xy}D_{yy}\frac{\partial P^*}{\partial y} - 0 + \frac{2}{\tau_c^2}D_{yy}\left(-D_{xx}\frac{\partial P^*}{\partial x} - D_{xy}\frac{\partial P^*}{\partial y}\right) \\ & +\frac{4}{\tau_c^2}D_{xy}\left(-D_{xy}\frac{\partial P^*}{\partial x} - D_{yy}\frac{\partial P^*}{\partial y}\right)\Bigg) \end{aligned} \tag{7.117}$$

$$= \frac{\tau_c}{3}\left(-\frac{3}{\tau_c^2}D_{xx}D_{yy}\frac{\partial P^*}{\partial x} - \frac{6}{\tau_c^2}D_{xy}D_{xy}\frac{\partial P^*}{\partial x} - \frac{9}{\tau_c^2}D_{xy}D_{yy}\frac{\partial P^*}{\partial y}\right) \tag{7.118}$$

$$= -\frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial P^*}{\partial x} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{xy}D_{yy}\frac{\partial P^*}{\partial y}. \tag{7.119}$$

The other components of $\boldsymbol{Q}$ can be gained analogously, whereby more details on the insertion of the terms can be found in appendix D.4:

$$Q_{113} = -\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial P^*}{\partial z}, \tag{7.120}$$

$$Q_{222} = -\frac{3}{\tau_c}D_{yy}D_{xy}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{yy}D_{yy}\frac{\partial P^*}{\partial y}, \tag{7.121}$$

$$Q_{123} = -\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial z}, \tag{7.122}$$

$$Q_{111} = -\frac{3}{\tau_c}D_{xx}D_{xx}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial y}, \tag{7.123}$$

$$Q_{112} = -\frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial P^*}{\partial y} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial P^*}{\partial y}, \tag{7.124}$$

$$Q_{233} = -\frac{1}{\tau_c} D_{xy} D_{zz} \frac{\partial P^*}{\partial x} - \frac{1}{\tau_c} D_{yy} D_{zz} \frac{\partial P^*}{\partial y}, \tag{7.125}$$

$$Q_{333} = -\frac{3}{\tau_c} D_{zz} D_{zz} \frac{\partial P^*}{\partial z}, \tag{7.126}$$

$$Q_{133} = -\frac{1}{\tau_c} D_{xx} D_{zz} \frac{\partial P^*}{\partial x} - \frac{1}{\tau_c} D_{xy} D_{zz} \frac{\partial P^*}{\partial y}, \tag{7.127}$$

$$Q_{223} = -\frac{1}{\tau_c} D_{yy} D_{zz} \frac{\partial P^*}{\partial z}. \tag{7.128}$$

Consequently, the $\boldsymbol{Q}$ scale $a^3/(\tau_c \tau_f^2)$ as supposed in equation 7.12.

The P equations

With knowledge of the first order (τ_c^{-1}) in $\boldsymbol{Q}$, 2 orders of $\boldsymbol{P}$ can be computed. Here, the procedure is shown exemplarily for P_{x1} from equation 7.81. Inserting the Q_{111}, Q_{211} and Q_{311} (setting o to x, y, z) into equation 7.81 yields:

$$\begin{aligned} P_{x1} = &-\frac{\tau_c}{2}\left(\frac{\partial}{\partial x}Q_{111} + \frac{\partial}{\partial y}Q_{211} + \frac{\partial}{\partial z}Q_{311} - B_{xx}P^*\right) \\ &+ \frac{\tau_c^2}{4}\left(\frac{\partial}{\partial t}\left(\frac{\partial}{\partial x}Q_{111} + \frac{\partial}{\partial y}Q_{211} + \frac{\partial}{\partial z}Q_{311} - B_{xx}P^*\right)\right. \\ &\left.+U_x^\infty \frac{\partial}{\partial x}\left(\frac{\partial}{\partial x}Q_{111} + \frac{\partial}{\partial y}Q_{211} + \frac{\partial}{\partial z}Q_{311} - B_{xx}P^*\right)\right) - \ldots \end{aligned} \tag{7.129}$$

$$= \frac{1}{\tau_c} D_{xx} P^* + D_{xx} D_{xx} \frac{\partial^2 P^*}{\partial x^2} + 2 D_{xx} D_{xy} \frac{\partial^2 P^*}{\partial x \partial y} + D_{xy} D_{xy} \frac{\partial^2 P^*}{\partial y^2} \tag{7.130}$$

Note that the time derivative in the second line and the U_x^∞ multiplication in the third line only are taken with respect to the P^*-term because this term dominates. For more information, see appendix D.5.
The solutions for the following $\boldsymbol{P}$ terms read:

$$P_{x2} = \frac{1}{\tau_c} D_{xy} P^* + D_{xx} D_{xy} \frac{\partial^2 P^*}{\partial x^2} + D_{xx} D_{yy} \frac{\partial^2 P^*}{\partial x \partial y} + D_{xy} D_{xy} \frac{\partial^2 P^*}{\partial x \partial y} + D_{xy} D_{yy} \frac{\partial^2 P^*}{\partial y^2}, \tag{7.131}$$

$$P_{x3} = D_{xx} D_{zz} \frac{\partial^2 P^*}{\partial x \partial z} + D_{xy} D_{zz} \frac{\partial^2 P^*}{\partial y \partial z}, \tag{7.132}$$

$$P_{y2} = \frac{1}{\tau_c} D_{yy} P^* + D_{xy} D_{xy} \frac{\partial^2 P^*}{\partial x^2} + 2 D_{xy} D_{yy} \frac{\partial^2 P^*}{\partial x \partial y} + D_{yy} D_{yy} \frac{\partial^2 P^*}{\partial y^2}, \tag{7.133}$$

$$P_{y3} = D_{xy} D_{zz} \frac{\partial^2 P^*}{\partial x \partial z} + D_{yy} D_{zz} \frac{\partial^2 P^*}{\partial y \partial z}, \tag{7.134}$$

$$P_{z3} = \frac{D_{zz}}{\tau_c} P^* + D_{zz} D_{zz} \frac{\partial^2 P^*}{\partial z^2}. \tag{7.135}$$

The j equations

Inserting the $\boldsymbol{P}$ components from equations 7.130-7.135 into the equations 7.72-7.75 for $\boldsymbol{j}$, all terms except the first order terms cancel out.
Even though, the equations from Wilemski (1976) are slightly different from the equations derived here, when inserting $\boldsymbol{U}^\infty$ in the final equation from (Wilemski 1976) for the external force field there, also his first order correction cancels out.
It seems that the amount of information available at this point does not suffice to find the next order term in the Fokker-Planck equation 7.19. From here, the approach of (Brinkman 1956), (Sack 1956), as described in (Risken 1989) is used.

7.4.4 Derivation of the telegraph equation

The procedure follows (Brinkman 1956) and (Sack 1956), and is also described in (Risken 1989). The $\boldsymbol{j}$ is eliminated out of the system. For that, the time derivative of equation 7.19 is taken:

$$\frac{\partial^2 P^*}{\partial t^2} = -U_x^\infty \frac{\partial^2 P^*}{\partial x \partial t} - \frac{\partial}{\partial t} \nabla \cdot \boldsymbol{j} \tag{7.136}$$

$$= -U_x^\infty \frac{\partial^2 P^*}{\partial x \partial t} - \nabla \cdot \frac{\partial \boldsymbol{j}}{\partial t} \tag{7.137}$$

In equation 7.137, $\boldsymbol{j}$ can be eliminated by inserting the moment equations from $\boldsymbol{j}$ from equations 7.23-7.25:

$$\begin{aligned}
\frac{\partial^2 P^*}{\partial t^2} &= -U_x^\infty \frac{\partial^2 P^*}{\partial x \partial t} - \frac{\partial}{\partial x}\left(-U_x^\infty \frac{\partial j_x}{\partial x} - \nabla \cdot \boldsymbol{P}_x - \frac{1}{\tau_c} j_x\right) \\
&\quad - \frac{\partial}{\partial y}\left(-U_x^\infty \frac{\partial j_y}{\partial x} - \nabla \cdot \boldsymbol{P}_y - \frac{1}{\tau_c} j_y\right) \\
&\quad - \frac{\partial}{\partial z}\left(-U_x^\infty \frac{\partial j_z}{\partial x} - \nabla \cdot \boldsymbol{P}_z - \frac{1}{\tau_c} j_z\right) \\
&= -U_x^\infty \frac{\partial^2 P^*}{\partial x \partial t} + \frac{\partial}{\partial x} \nabla \cdot \boldsymbol{P}_x + \frac{\partial}{\partial y} \nabla \cdot \boldsymbol{P}_y + \frac{\partial}{\partial z} \nabla \cdot \boldsymbol{P}_z \\
&\quad + \nabla \cdot \left(U_x^\infty \frac{\partial}{\partial x} \boldsymbol{j}\right) + \frac{1}{\tau_c} \nabla \cdot \boldsymbol{j} \\
&= -U_x^\infty \frac{\partial^2 P^*}{\partial x \partial t} + \frac{\partial}{\partial x} \nabla \cdot \boldsymbol{P}_x + \frac{\partial}{\partial y} \nabla \cdot \boldsymbol{P}_y + \frac{\partial}{\partial z} \nabla \cdot \boldsymbol{P}_z \\
&\quad + \left(U_x^\infty \frac{\partial}{\partial x}\right) \nabla \cdot \boldsymbol{j} + \frac{\partial U_x^\infty}{\partial y} \frac{\partial j_y}{\partial x} + \frac{1}{\tau_c} \nabla \cdot \boldsymbol{j} \qquad (7.138) \\
&= -U_x^\infty \frac{\partial^2 P^*}{\partial x \partial t} + \frac{\partial}{\partial x} \nabla \cdot \boldsymbol{P}_x + \frac{\partial}{\partial y} \nabla \cdot \boldsymbol{P}_y + \frac{\partial}{\partial z} \nabla \cdot \boldsymbol{P}_z \\
&\quad + \left(U_x^\infty \frac{\partial}{\partial x}\right) \nabla \cdot \boldsymbol{j} + \dot{\gamma}\left(D_{xy} \frac{\partial^2 P^*}{\partial x^2} + D_{yy} \frac{\partial^2 P^*}{\partial x \partial y}\right) + \frac{1}{\tau_c} \nabla \cdot \boldsymbol{j}. \qquad (7.139)
\end{aligned}$$

In equation 7.138 the product rule requires the separate derivation of U_x^∞ with respect to y. Then, another j_y enters which has been replaced by the lowest order of j_y^h from equation 7.114. Equation 7.139 also contains $\boldsymbol{j}$ again. To eliminate $\boldsymbol{j}$, the starting equation for P^*, i.e. equation 7.19, is transformed to an equation for $\boldsymbol{j}$:

$$\nabla \cdot \boldsymbol{j} = -U_x^\infty \frac{\partial P^*}{\partial x} - \frac{\partial P^*}{\partial t}. \tag{7.140}$$

After multiplying both sides of the equation with τ_c and inserting the terms for $\boldsymbol{P}$ from equations 7.130-7.135, the result is:

$$\begin{aligned}
&\tau_c \frac{\partial^2 P^*}{\partial t^2} + \frac{\partial P^*}{\partial t} \\
&= -U_x^\infty \frac{\partial P^*}{\partial x} - 2\tau_c U_x^\infty \frac{\partial^2 P^*}{\partial x \partial t} - \tau_c U_x^\infty U_x^\infty \frac{\partial^2 P^*}{\partial x^2} \\
&\quad + D_{xx} \frac{\partial^2 P^*}{\partial x \partial x} + 2D_{xy} \frac{\partial^2 P^*}{\partial x \partial y} + D_{yy} \frac{\partial^2 P^*}{\partial y^2} + D_{zz} \frac{\partial^2 P^*}{\partial z^2} \\
&\quad + \tau_c \left(D_{xx}^2 \frac{\partial^4 P^*}{\partial x^4} + 2D_{xx} D_{xy} \frac{\partial^4 P^*}{\partial x^3 \partial y} + D_{xy}^2 \frac{\partial^4 P^*}{\partial x^2 \partial y^2} \right) \\
&\quad + 2\tau_c \left(D_{xx} D_{xy} \frac{\partial^4 P^*}{\partial x^3 \partial y} + D_{xx} D_{yy} \frac{\partial^4 P^*}{\partial x^2 \partial y^2} + D_{xy}^2 \frac{\partial^4 P^*}{\partial x^2 \partial y^2} + D_{xy} D_{yy} \frac{\partial^4 P^*}{\partial x \partial y^3} \right) \\
&\quad + 2\tau_c \left(D_{xx} D_{zz} \frac{\partial^4 P^*}{\partial x^2 \partial z^2} + D_{xy} D_{zz} \frac{\partial^4 P^*}{\partial x \partial y \partial z^2} \right) \\
&\quad + \tau_c \left(D_{xy}^2 \frac{\partial^4 P^*}{\partial x^2 \partial y^2} + 2D_{xy} D_{yy} \frac{\partial^4 P^*}{\partial x \partial y^3} + D_{yy}^2 \frac{\partial^4 P^*}{\partial y^4} \right) \\
&\quad + 2\tau_c \left(D_{xy} D_{zz} \frac{\partial^4 P^*}{\partial x \partial y \partial z^2} + D_{yy} D_{zz} \frac{\partial^4 P^*}{\partial y^2 \partial z^2} \right) \\
&\quad + \tau_c \left(D_{zz}^2 \frac{\partial^4 P^*}{\partial z^4} \right) - \tau_c \dot{\gamma} \left(D_{xy} \frac{\partial^2 P^*}{\partial x^2} + D_{yy} \frac{\partial^2 P^*}{\partial x \partial y} \right).
\end{aligned} \tag{7.141}$$

7.5 Conclusion

For $\tau_c \to 0$, equation 7.141 reduces to the Fokker-Planck equation of Breedveld et al. (1998) and Sierou & Brady (2004), cf. equation 6.1:

$$\frac{\partial P}{\partial t} = -y\dot{\gamma} \frac{\partial P}{\partial x} + D_{xx}(t) \frac{\partial^2 P}{\partial x^2} + D_{yy}(t) \frac{\partial^2 P}{\partial y^2} + D_{zz}(t) \frac{\partial^2 P}{\partial z^2} + 2D_{xy}(t) \frac{\partial^2 P}{\partial x \partial y}. \tag{7.142}$$

Note that the diffusion coefficients from Sierou and Brady become constant for the diffusive regime (Sierou & Brady 2004) and this is exactly what shall be regarded here. Breedveld et al. (1998) derived the same equation as 7.142 with constant diffusion coefficients. Most important, the correlation time of the colored-noise velocity is not approaching 0. In fact, it is comparable to τ_f, whereas the analysis throughout this chapter is based on the assumption that $\tau_c \ll \tau_f$ but non-white. Without the

assumption that $\tau_c \ll \tau_f$, the series expansion in equation 7.141 would require an infinite number of terms.

As mentioned above in section 6.3.1, the solution P_2 fulfills the colored-noise Fokker-Planck equation 6.51 with an error of zero (even though, the exponential terms were neglected in the derivation). In the present chapter, P^* is regarded which results from an integration of P_2 over velocity space, (equation 7.2). Inserting P^* into the Fokker-Planck equation from (Breedveld et al. 1998), i.e. equation 7.142 with constant diffusion coefficients, results in an error of $O(\tau_c)$. Inserting P^* into the new reduced position-space Fokker-Planck equation 7.141 yields an error of $O(\tau_c^2)$. Consequently, based on the colored-noise Fokker-Planck equation, in the reduced form one order in τ_c is gained. The calculations of the error have been performed with Maple$^{\text{TM}}$18.

7.6 Short summary of chapter 7

The colored-noise Fokker-Planck equation derived in chapter 6 is reduced to an equation in position space and compared to the traditionally used equations in position space. It is shown that the traditional equations are recovered for a velocity autocorrelation time approaching zero. For short velocity correlation times, additional correction terms are presented. For considerable velocity correlation times, the position formulation requires an infinite number of correction terms. This gives rise to the assumption that consequently, a colored-noise Fokker-Planck equation as presented in chapter 6 is necessary.

8 Outlook and summary

In the preceding chapters, an alternative Fokker-Planck formulation for non-Brownian particles in shear flow was derived, namely a colored-noise Fokker-Planck formulation. This included a model for the hydrodynamic velocity component. Further, this colored-noise Fokker-Planck equation was reduced to a new position-space formulation with second order time derivative and higher order moment components. This chapter shall give an outlook to important aspects arising out of the present work. Firstly, a short literature overview on simulations in the context of non-Brownian suspensions is given. Further, exemplary simulation results gained from a code called *Bluebottle* (Sierakowski & Prosperetti status January 23rd 2015) are shown. Possible extensions of the new Fokker-Planck model are presented. Finally, a summary is given.

8.1 Basic introduction to the numerical simulations of shear-induced diffusion

Here shall be given an insight into the numerical side of the problem of shear-induced diffusion of non-Brownian particles. The purpose of this section is to give a short literature survey of simulations that have been done in that context. The coefficients for the new Fokker-Planck model 6.50 have to be gained either from experimental data or via numerical simulation results. As can be seen from equation 6.44, 6.47 and 6.49, the B_{ij} coefficients are related to the mean square displacements through D_{ij}. The mean square displacements have been measured in various situations, e.g. numerically by Sierou & Brady (2004), or experimentally by Breedveld et al. (1998).

8.1.1 Small literature overview to numerical simulations

There are various simulation techniques, for example Alder & Wainwright (1959), (Alder & Wainwright 1967), (Alder & Wainwright 1970) have used molecular dynamics approaches for the study of molecules. Their simulation results are important in the context of section 3.1.3.1 and the following section 8.2.1. On the level of Brownian and non-Brownian particles, a central method of simulation for low Reynolds number flows which has been mentioned several times throughout the present work is the Stokesian dynamics method (Brady & Bossis 1988), cf. section 3.2. Brady and Bossis say that their method includes the Brownian dynamcis method, see e.g. (Ermak & McCammon 1978), as a special case. The derivation of the hydrodynamic interactions plays an important role in Stokesian dynamics, which was also mentioned in section 3.2.1. The Stokesian dynamics method has been used by various authors also in the context of shear-induced diffusion, e.g. in the work of Marchioro & Acrivos (2001) who

studied non-Brownian particles in shear-induced diffusion and analyzed the Gaussian distribution of the displacements in y- and z-direction. This will be referred to in the subsequent section 8.2.2 again. Further, Marchioro and Acrivos report a very interesting phenomenon, namely negative correlation of velocities in y- and z-directions. In this context, Drazer et al. (2002) also used Stokesian dynamics analyzing negative correlations of velocities in y- and z-directions with regard to particle volume fractions, where it turned out that this phenomenon especially occurs for low volume fractions, cf. also (Drazer, Koplik, Kushid & Acrivos 2004). This will be further discussed in section 8.2.1. Sierou & Brady (2001) have refined the many-body long-range interaction of the Stokesian dynamics method to better account for higher numbers of particles. This method is named accelerated Stokesian dynamics method and has been widely used by Sierou & Brady (2004) in the context of shear-induced diffusion, see also overview in section 8.1.1.1. Further, the work of Santamaría-Holek et al. (2009b) ought to be mentioned in this context, see also sections 2.3.2 and 6.1.1. They studied the validity of the fluctuation dissipation theorem for non-Brownian particles and performed numerical simulations with the lattice Boltzmann method (see (Ladd 1994a), (Ladd 1994b)) of the effective diffusivity dependent on particle volume fraction and Reynolds number. Other simulation techniques such as dissipative particle dynamics (DPD), see (Hoogerbrugge & Koelman 1992), are not considered in the present work.

8.1.1.1 Preliminary considerations

Some preliminary considerations for the numerical simulation of shear-induced diffusion have to be taken into account. Sierou & Brady (2004) give explicit information on the appropriate number of particles, particle volume fraction ϕ, shear rate $\dot{\gamma}$, box size measurements. An infinitely large domain cannot be simulated so in case of periodic boundary conditions the box measurements and particle volume fractions have to ensure that a particle re-entering on the other side of the box is not correlated with itself (Sierou & Brady 2004). Further, Sierou & Brady (2004) show measurements with a fixed particle volume fraction where the number of particles is increased (and thus the box size to keep ϕ constant). The results show that the diffusivity increases with higher number of particles.

8.1.2 The used code *Bluebottle*

The simulation results that are presented in this chapter in figures 8.1a-8.4b have been performed by a code called *Bluebottle* (Sierakowski & Prosperetti status January 23rd 2015). *Bluebottle* is a research code and has been developed by Adam Sierakowski[1] under supervision of Andrea Prosperetti from the Johns Hopkins University in Baltimore, Maryland, USA. First results from simulations with *Bluebottle* concerning shear-induced diffusion have also been presented (Lukassen, Sierakowski & Oberlack 2014). For the present work, the code has been used in a pre-release version of the recently

[1] `http://lucan.me.jhu.edu/wiki/index.php/Adam_Sierakowski`, status January, 23 2015

released version (available via GitHub under the Apache Licence Version 2.0)[2]. The following paragraph is based on the information from (Sierakowski & Prosperetti status January 23rd 2015).

Bluebottle is a GPU-centric finite-difference incompressible Navier-Stokes flow solver coupled with a particle method which fully resolves spherical particles. The underlying method for particles is the Physalis method which presumes Stokes flow in close proximity of particles due to the no-slip condition, see (Zhang & Prosperetti 2005), (Gudmundsson & Prosperetti 2013). The Physalis method takes benefit out of the existence of an analytical solution of Stokes flow around spherical particles. Due to this, the Physalis method is restricted to spherical particles and thus, also the *Bluebottle* implementation is restricted to spherical particles[3]. However, this does not pose a limitation for the present work since here, only spherical particles are regarded. The implementation is capable of treating low to moderate Reynolds number. The solution procedure of *Bluebottle* contains an iteration procedure between the flow and the particle solution which gave rise to a parallel GPU implementation[3]. The analytical solution of the Stokes flow is done in terms of so-called spherical harmonics and requires the computation of several coefficients, see also (Kim & Karrila 2005, Sect. 4.2), (Lamb 1962). These coefficients are computed iteratively and adjusted to the flow solver. In this sense, it is not necessary to conform the grid to the spherical shape of the particles which is an advantage of the Physalis method. The implementation also considers lubrication which, as already mentioned in section 2.2.1, is an important phenomenon in the context of hydrodynamic interaction.

Technical components

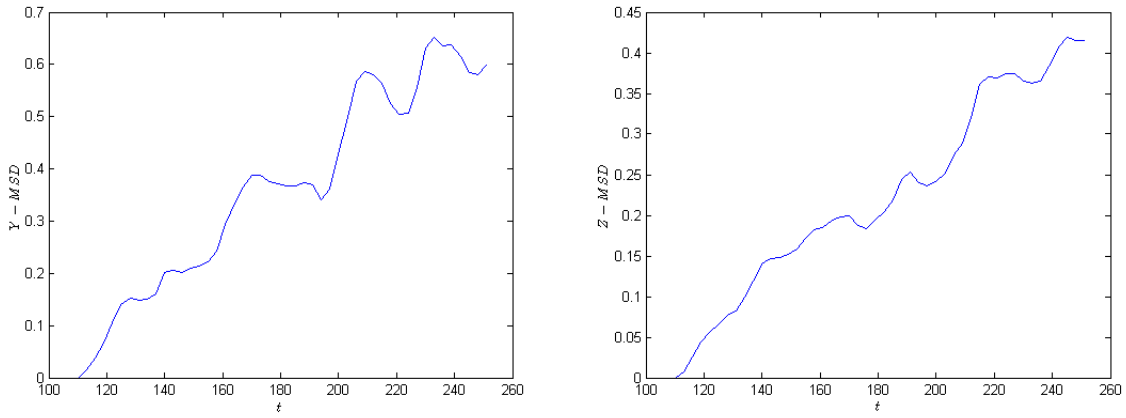
The simulations for the present work have been running on the CUDA cluster of the Graduate School of Excellence Computational Engineering at TU Darmstadt. For the analysis of the results, Matlab® (R2011a) (The MathWorks®, Inc.) is used. Matlab® routines such as reading out the current particle positions and velocities have been used from the *Bluebottle* framework and have been extended to measure mean square displacements (which account for the walls in y-directions and re-entering particles into the domain in z-direction) and velocity correlations in time. The visualization is done by the Matlab® plotting tools.

8.1.3 The test case

The *Bluebottle* framework enables to set several simulation parameters, the parameters which are of most interest for the present work are given below. In the simulations, volume fractions of $\phi = 0.121$ (50 particles, case 1) and $\phi = 0.249$ (103 particles, case 2) were used. The box size ranges were of -6 to 6 in x, y, z-directions, resulting in a

[2] `https://github.com/groundcherry/bluebottle`, status January, 23 2015

[3] `http://lucan.me.jhu.edu/wiki/index.php/The_Physalis_method`, status January, 23 2015



(a) Mean square displacement in y-direction (b) Mean square displacement in z-direction

Figure 8.1: Mean square displacements for case 2 (103 particles)

volume of 12^3. The density of the particles ρ_p, the fluid density ρ_f, the viscosity η, the radius a are given in dimensionless form according to:

$$\rho_p = 1, \qquad \rho_f = 1, \qquad \eta = 1, \qquad a = 1. \tag{8.1}$$

There is applied shear flow in x-direction. In y-direction, there are contrary moving walls with velocity ± 1. The resulting shear rate is $\dot{\gamma} = 1/6$. x and z-direction have periodic boundary conditions.

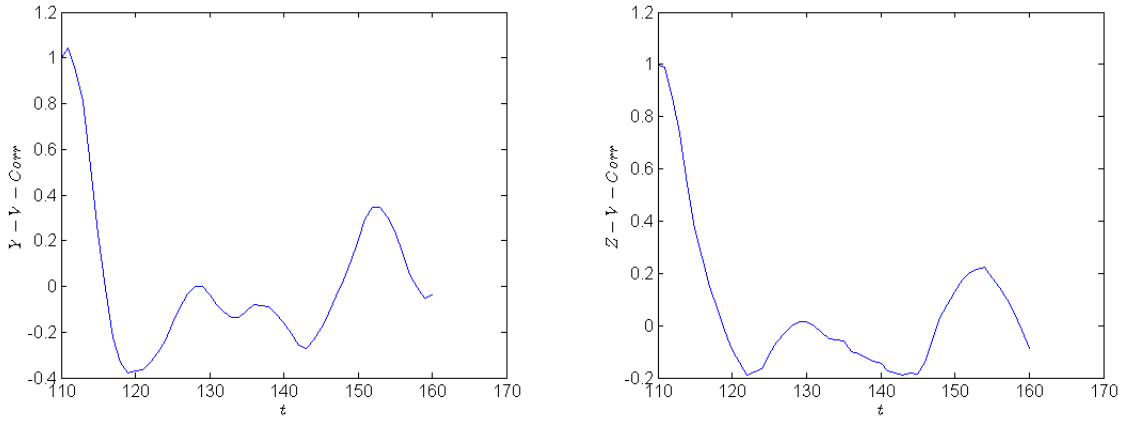
The figures 8.1a and 8.1b show the mean square displacements in y- and z-direction of case 2. The figures show the linearly growing mean square displacements as indicated in equations 2.28 and 2.29. In x-direction, the mean square displacements have different behavior due to the underlying shear flow.

8.2 Rise to future work

Throughout this work, a lot of effects have been summarized. Some of them give rise to future research and are worth to be further investigated in the context of the colored-noise velocity model (section 6.2), the colored-noise Fokker-Planck equation 6.50 and the reduced form 7.141.

8.2.1 Extension of the colored-noise velocity model

A very important issue in modeling the behavior of particles is their long time behavior. In this context, there was observed that Brownian particles exhibit the so-called long-time tail behavior, see e.g. (Dhont 1996). In section 3.1.3.1 was introduced the conflict considering the use of Stokes law in the equation of motion for the Brownian particle. This conflict was substantiated by the numerical simulation results of Alder &

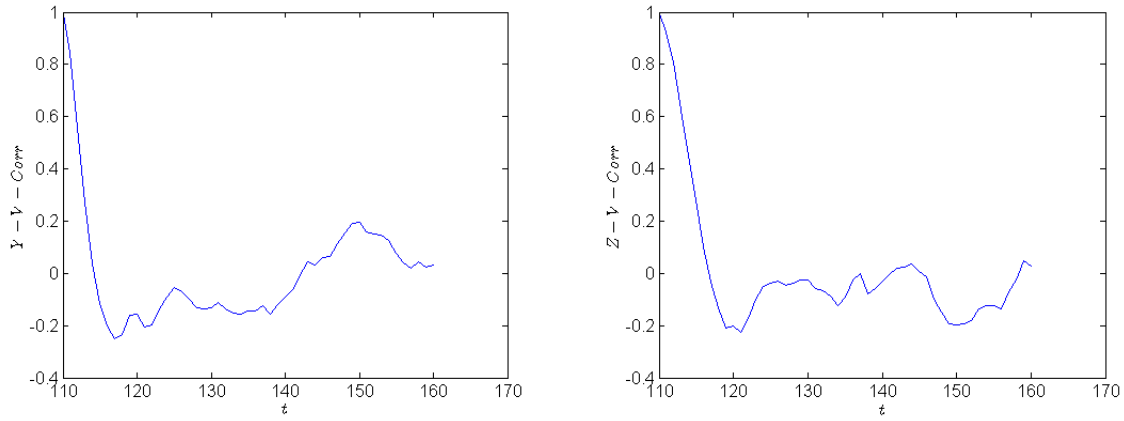


(a) y-direction, 50 particles (b) z-direction, 50 particles

Figure 8.2: Velocity autocorrelation for y- and z-direction for case 1

Wainwright (1970). They detected that the velocity autocorrelation decays rather with $t^{-3/2}$ for long times than exponentially, thus that the correlation persists longer. This was explained by a backflow, see (Clerx & Schram 1992). Altogether such behavior is called long-time tail behavior. This gave rise to an extension of the equation of motion (the velocity equation) for Brownian particles including added mass and history terms (equation 3.31) (Russel et al. 1991). They found that the velocity autocorrelation derived from the incorporation of added mass and history terms decays with $t^{-3/2}$ for long times. In the present work, the Stokes flow solution is not used for the computation of the coefficients of the colored-noise model, in this way, the conflict does not influence the used model. Still, the autocorrelation of the hydrodynamic velocity may also contain a deviation from pure exponential decay, so it may be worth considering the incorporation of similar effects such as added mass and history terms.

In addition to a slow $t^{-3/2}$-decay, simulations, e.g. by Rahman (1964), Rahman (1966), show a negative velocity autocorrelation for intermediate times, cf. section 3.1.3.1. This was also substantiated by Dhont (1996), that the Brownian particle leaving the surrounding particles, will reverse its velocity in between.
In non-Brownian particle simulation with Stokesian dynamics has been found a comparable behavior by Marchioro & Acrivos (2001) and Drazer et al. (2002). Drazer et al. (2002) showed that especially for small particle volume fractions the negative velocity autocorrelation emerges more than for higher particle volume fractions. Due to Drazer et al., this may be due to more two-particle interactions in lower volume fractions which cause reverse velocities when particles come into contact which is not happening in many-particle interactions. Drazer et al. show that this effect decreases substantially for higher concentrations or volume fractions. As the diffusion coefficient can be written in terms of an integral of the velocity autocorrelation function (equation 6.43) this negative contribution in the velocity autocorrelation function decreases the diffusion coefficient (Marchioro & Acrivos 2001).
This negative velocity correlation can be also shown by simulations with *Bluebottle*. In figures 8.2a-8.3b, the negative velocity correlation can be found. Further, the effect of



(a) y-direction, 103 particles (b) z-direction, 103 particles

Figure 8.3: Velocity autocorrelation for y- and z-direction for case 2

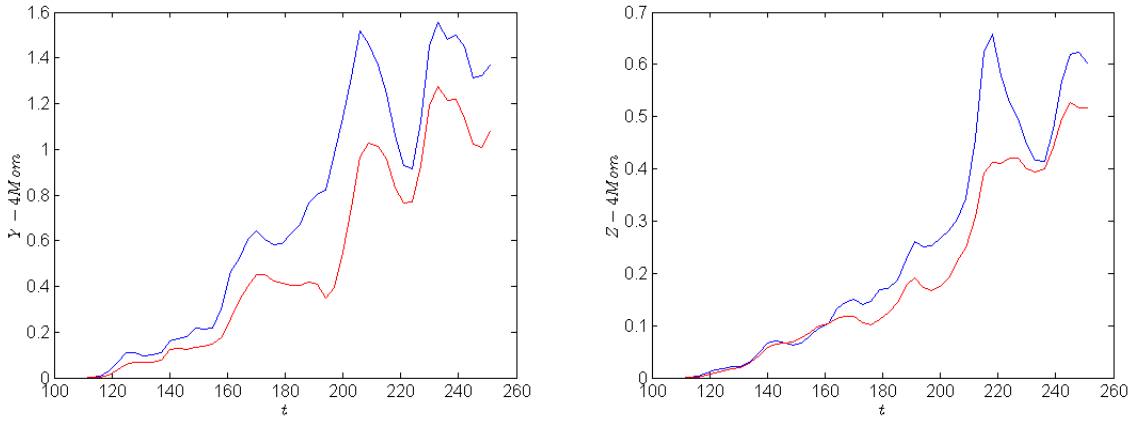
negative velocity autocorrelation is more significant for the smaller volume fraction with 50 particles, as also indicated by Drazer et al. (2002).
The exponential colored-noise velocity model used in the present work cannot reproduce such negative effects arising in lower volume fractions. So, as a future work, it is promising, besides a possible extension with added mass and history terms in general, to consider approaches such as presented in (Zwanzig & Bixon 1970) and (Berne et al. 1966) which reproduce negative correlation times.

8.2.1.1 Reduction to position space

A reduction to position space in terms of the approaches, mentioned in sections 6.1.1 and 7.1.1, which result in a time dependent diffusion coefficient by incorporating memory in the diffusion, are promising, especially as shown in (San Miguel & Sancho 1980) and the matrix continued fraction (Risken 1989). But also for these approaches, a model for the velocity autocorrelation is necessary where either the Ornstein-Uhlenbeck model used in the present work could be used or an extension as outlined above. Memory functions have also been considered by Dhont (1996). Special forms of Fokker-Planck equations, also referred to as generalized Fokker-Planck equations, can be derived, e.g. (Adelman 1976), (Wilemski 1976, Appendix).

8.2.2 Fourth order moments

The reduced form of the colored-noise Fokker-Planck equation, i.e. equation 7.141, exhibits higher order derivatives in position space besides the second order. The resulting form is similar to the Kramers-Moyal expansion series (mentioned in section 4.2.2, equation 4.39), except for the second order time derivative. From Pawula (1967) it is known that the expansion may either be interrupted after the second term or not at all. Consequently, for the present case, it is only possible to stop the expansion under



(a) 4-th moment in y-direction

(b) 4-th moment in z-direction

Figure 8.4: Fourth order moments for y- and z-direction, case 2 (103 particles)

the assumption that τ_c is small enough. However, the fourth order components are not exactly the fourth order moments. A resulting question is if it would be possible, to eliminate the second order time derivative and create terms corresponding to the fourth order moments of the position variables. Consequently, it may be worth investigating the influence of higher moments. For a Gaussian distributed position variable, the fourth cumulant is zero (equation 4.13, cf. (Gardiner 2009)). The consequence is that $\langle y^4 \rangle = 3\langle y^2 \rangle$, the same applies for z. Also in literature, it has already been shown that the distribution is indeed Gaussian, see (Breedveld et al. 1998). According to Marchioro & Acrivos (2001), this can be confirmed by calculating higher order moments. Simulations using *Bluebottle* can also show the same effect when comparing the fourth order moment with 3 times the second moment squared: Figure 8.4a and 8.4b show the fourth moment in y- and z-direction respectively (blue plot), compared to the fourth order moment in terms of 3 times the second order moment squared (red plot) (i.e. by use of the cumulant rule (equation 4.13)).

8.2.3 Incorporating the particle volume fraction and effective viscosity

An important influence factor is the particle volume fraction. For increasing particle volume fraction, the diffusivity increases, see (Sierou & Brady 2004), i.e. the diffusion coefficients can be written as $\boldsymbol{D} = \dot{\gamma} a^2 \tilde{\boldsymbol{D}}(\phi)$ (Breedveld 2000), (Breedveld et al. 2002), (Morris & Brady 1996), whereas the increase stops at higher volume fractions (Breedveld, van den Ende, Bosscher & Jongschaap 2001a). This aspect is included in the colored-noise Fokker-Planck equation 6.50 since the B_{ij} coefficients are gained via the mean square displacements.
Further, it can be shown that also the correlation time depends on the particle volume fraction. The time scale for correlations becomes shorter, the more interactions take place, i.e. the higher the particle volume fraction (Drazer et al. 2002), cf. 2.3.2. Additionally, Drazer et al. (2002) also show that the velocity autocorrelation time differs

from y- to z-direction, which is not included in the model in the present work so far. From Dhont (1996) can be found that the effective viscosity can be used to determine the long-time diffusion coefficient of Brownian particles.

The open question concerning the colored-noise Fokker-Planck equation 6.50 is, if it is possible to also incorporate the volume fraction dependence into the colored-noise correlation time, i.e. use a modified coefficient in equation 6.50, instead of $1/\tau_f$, some relation $\tau_c = f(\phi) \cdot \tau_f$. It could be considered to relate $f(\phi)$ to the effective viscosity, in analogy to the Brownian friction which can be written as an effective friction (Dhont 1996). For more information on influence factors also on diffusion coefficients, see (Foss & Brady 1999).

8.3 Summary

Within this work, a framework for non-Brownian particles in shear flow has been presented. It is partly based on (Lukassen 2012), and more important on (Lukassen & Oberlack 2014b). The long correlation times of shear-induced diffusion require a special treatment in the corresponding stochastic description. In this context, a multiple time scale analysis was introduced to expose the difference to the description of Brownian particles and more important to analyze non-Brownian particles with regard to the Markov process assumption. In the context of this time scale analysis, it is pointed out that the special case of the treatment of external force fields for Brownian particles shows similarities to the shear-induced diffusion process. For Brownian particles this yields the Kramers equation, a Fokker-Planck equation in position velocity space (van Kampen 2007). In the present work, it is assumed that the description of non-Brownian particles also may require a coupled description which was substantiated by the fact that the position of non-Brownian particles in shear flow alone is no longer Markovian. Instead, a coupled variable of position and a colored-noise velocity variable can recover the Markov property of the problem. The colored-noise velocity is the velocity component due to hydrodynamic interactions with surrounding particles. This was modeled as an Ornstein-Uhlenbeck process. Based on that, a colored-noise Fokker-Planck equation was derived to describe non-Brownian particles in shear flow. A Gaussian solution based on the algorithm from (van Kampen 2007) was derived and compared to the Gaussian solution of the models used traditionally in this context, as given in (Breedveld et al. 1998). Furthermore, the colored-noise Fokker-Planck equation was reduced to position space by combining the approaches of (Wilemski 1976) and (Brinkman 1956), (Sack 1956). The reduction is based on an expansion series in the correlation time of the colored-noise. When compared to the traditionally used position-space Fokker-Planck equations (Breedveld et al. 1998), (Sierou & Brady 2004), the new reduced position-space formulation exhibits correction terms in higher orders of the correlation time. Especially, the new position-space formulation exhibits second order in time derivatives and also higher order spatial derivatives. Only for reducing the colored-noise correlation time to a white noise, the traditional Fokker-Planck equations are recovered. Finally, an extended

outlook was given to show possible extensions for the models introduced within this work. This was substantiated by exemplary simulation results.

9 Bibliography

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A Appendix 1

Here will be shown the $(\triangle t)^2$ dependence of $(\triangle x)^2$ in analog form to equation 3.24 from (Risken 1989).

$$\begin{aligned}\int_{t_n}^{t}\int_{t_n}^{t}\langle V_i(t')V_j(t'')\rangle_{\boldsymbol{V}}dt'dt'' = V_i(t_n)V_j(t_n)\tau_f^2\left(1-\exp\left(\frac{-(t-t_n)}{\tau_f}\right)\right)^2 \\ + B_{ij}\tau_f\left(\tau_f(t-t_n)+\tau_f^2\exp\left(\frac{-(t-t_n)}{\tau_f}\right)-\tau_f^2\right) \\ - \frac{1}{2}B_{ij}\tau_f^3\left(1-\exp\left(\frac{-(t-t_n)}{\tau_f}\right)\right)^2.\end{aligned} \tag{A.1}$$

For $t = t_{n+1}$, in the limit $\triangle t = (t_{n+1} - t_n) \to 0$ follows: $\int_{t_n}^{t}\int_{t_n}^{t}\langle V_i(t')V_j(t'')\rangle_{\boldsymbol{V}}dt'dt'' = V_i(t_n)V_j(t_n)(\triangle t)^2$. This was also included in (Lukassen & Oberlack 2014b).

B Appendix 2

This appendix is taken from (Lukassen & Oberlack 2014b). The following rule from (Risken 1989) for a general equation for the velocity autocorrelation according to an Ornstein-Uhlenbeck process is transferred to the colored-noise velocity:

$$\langle V_i(t')V_j(t'')\rangle_{\boldsymbol{V}} = V_i(0)V_j(0)\exp\left(\frac{-(t'+t'')}{\tau_f}\right) + \frac{B_{ij}\tau_f}{2}\left\{\exp\left(\frac{-|t'-t''|}{\tau_f}\right) - \exp\left(\frac{-(t'+t'')}{\tau_f}\right)\right\}. \tag{B.1}$$

Transferring equation (B.1) to $\langle V_i(t')V_j(t'')\rangle_{\boldsymbol{V}}$ yields:

$$\langle V_i(t')V_j(t'')\rangle_{\boldsymbol{V}} = V_i(t_n)V_j(t_n)\exp\left(\frac{-(t'+t''-2t_n)}{\tau_f}\right) + \frac{B_{ij}\tau_f}{2}\left\{\exp\left(\frac{-|t''-t'|}{\tau_f}\right) - \exp\left(\frac{-(t'+t''-2t_n)}{\tau_f}\right)\right\}. \tag{B.2}$$

It follows:

$$\begin{aligned}\langle \triangle V_i \triangle V_j\rangle_{\boldsymbol{V}} &= V_i(t_n)V_j(t_n)\exp(-2\triangle t/\tau_f) + \frac{B_{ij}\tau_f}{2}\left(\exp(0) - \exp(-2\triangle t/\tau_f)\right) \\ &- 2\left\{V_i(t_n)V_j(t_n)\exp(-\triangle t/\tau_f) + \frac{B_{ij}\tau_f}{2}\left(\exp(-\triangle t/\tau_f) - \exp(-\triangle t/\tau_f)\right)\right\} \\ &+ V_i(t_n)V_j(t_n) \\ &= V_i(t_n)V_j(t_n)\exp(-2\triangle t/\tau_f) + \frac{B_{ij}\tau_f}{2}\left(1 - \exp(-2\triangle t/\tau_f)\right) \\ &- 2V_i(t_n)V_j(t_n)\exp(-\triangle t/\tau_f) + V_i(t_n)V_j(t_n).\end{aligned} \tag{B.3}$$

For $\triangle t \to \infty$ applies:

$$\langle \triangle V_i \triangle V_j\rangle_{\boldsymbol{V}} = \frac{B_{ij}\tau_f}{2} + V_i(t_n)V_j(t_n). \tag{B.4}$$

In case that the $\langle\rangle$-brackets are related to $t = 0$ (instead of $\boldsymbol{V}$ in the index) with $t > \tau_f$, $t \to \infty$, it follows $\langle V_i(t)V_j(t)\rangle = B_{ij}\tau_f/2$.

For $\triangle t \to 0$ applies:

$$\langle \triangle V_i \triangle V_j\rangle_{\boldsymbol{V}} = B_{ij}\triangle t + O((\triangle t)^2). \tag{B.5}$$

C Appendix 3

In the following, $\int_0^\infty \lim_{t\to\infty}\langle V_x(t+t')V_x(t)\rangle dt'$ is defined in order to get B_{xx}, taken from (Lukassen & Oberlack 2014b).
It will be shown the relation 6.46, i.e. $\int_0^t \langle V_x(t+t')V_x(t)\rangle dt' = \frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle$ in the limit $t\to\infty$. The case for B_{xy} is analog.
D_{xx} can be written as shown in equation 2.31: $\partial_t\langle xx\rangle - 2\langle x\,(\dot{\gamma}y)\rangle = 2D_{xx}$.
With $x(t) = \int_0^t (V_x(t') + U_x^\infty(t'))dt'$, $x(0) = 0$, it can be found:

$$\langle xx\rangle = \left\langle \left(\int_0^t (V_x(t') + U_x^\infty(t'))dt'\right)^2 \right\rangle, \tag{C.1}$$

$$\partial_t\langle xx\rangle = 2\left\langle \left(\int_0^t (V_x(t') + U_x^\infty(t'))dt'\right)\,(V_x(t) + U_x^\infty(t))\right\rangle, \tag{C.2}$$

with $\left\langle \int_0^t V_x(t)V_x(t')dt'\right\rangle = \left\langle \int_0^t V_x(t)V_x(t+t')dt'\right\rangle$ as $t\to\infty$. This can be shown by integrating equation 6.16 for both cases, t' and $t+t'$.

Inserting this into the equation for D_{xx} yields:

$$\begin{aligned}
D_{xx} = \frac{1}{2}\frac{\partial}{\partial t}\langle xx\rangle - \langle x(\dot{\gamma}y)\rangle = \\
&\left\langle \int_0^t V_x(t)V_x(t+t')dt'\right\rangle + \left\langle \int_0^t U_x^\infty(t)V_x(t+t')dt'\right\rangle \\
&+ \left\langle \int_0^t V_x(t)U_x^\infty(t')dt'\right\rangle + \left\langle \int_0^t U_x^\infty(t)U_x^\infty(t')dt'\right\rangle \\
&- \left\langle \int_0^t U_x^\infty(t)V_x(t+t')dt'\right\rangle - \left\langle \int_0^t U_x^\infty(t)U_x^\infty(t')dt'\right\rangle, \\
D_{xx} = \left\langle \int_0^t V_x(t)V_x(t+t')dt'\right\rangle &+ \left\langle \int_0^t V_x(t)U_x^\infty(t')dt'\right\rangle.
\end{aligned} \tag{C.3}$$

Thus:

$$\left\langle \int_0^t V_x(t)V_x(t+t')dt'\right\rangle = D_{xx} - \left\langle \int_0^t V_x(t)U_x^\infty(t')dt'\right\rangle. \tag{C.4}$$

It can be shown: $\langle\int_0^t V_x(t)U_x^\infty(t')dt'\rangle = \langle V_x(t)\int_0^t U_x^\infty(t')dt'\rangle$.
So it follows: $\langle\int_0^t V_x(t)U_x^\infty(t')dt'\rangle = \left\langle \frac{dx^h}{dt}\int_0^t \dot{\gamma}y(t')dt'\right\rangle$. With the diffusion coefficient in x-direction from (Sierou & Brady 2004), see also equation 6.6,

$$D_{xx}(t) = \frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle + \left\langle \frac{dx^h}{dt}\int_0^t \dot{\gamma}y(t')dt'\right\rangle, \tag{C.5}$$

equation C.4 yields:

$$\Big\langle \int_0^t V_x(t)V_x(t+t')dt' \Big\rangle = \int_0^t \langle V_x(t)V_x(t+t')\rangle dt' = \frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle. \tag{C.6}$$

D More details on the equations for the reduced form

In analog form to the integration of the $\boldsymbol{j}$ terms, here will be shown the integration of the $\boldsymbol{P}$, $\boldsymbol{Q}$, and $\boldsymbol{R}$-terms by use of the same procedure of integration as in (Wilemski 1976). In accordance with the integration of $\boldsymbol{j}$, the initial terms (i.t. in equation 7.71) are omitted also in the following integrations.

D.1 Integration of P_{x1}

$$
\begin{aligned}
P_{x1} &= -\int_0^{t_*} \exp\left(\frac{-2(t_*-\tau)}{\tau_c}\right)\left(\nabla_*\cdot\boldsymbol{Q}_{o11} - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}Q_{211} - B_{xx}P^*\right)d\tau \\
&= -\left[\frac{\tau_c}{2}\left(\nabla_*\cdot\boldsymbol{Q}_{o11} - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}Q_{211} - B_{xx}P^*\right)\right. \\
&\quad \left. -\int_0^{t_*}\frac{\tau_c}{2}\exp\left(\frac{-2(t_*-\tau)}{\tau_c}\right)\frac{\partial}{\partial\tau}\left(\nabla_*\cdot\boldsymbol{Q}_{o11} - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}Q_{211} - B_{xx}P^*\right)d\tau\right] \\
&= -\left[\frac{\tau_c}{2}\left(\nabla_*\cdot\boldsymbol{Q}_{o11} - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}Q_{211} - B_{xx}P^*\right)\right. \\
&\quad -\frac{\tau_c^2}{4}\frac{\partial}{\partial t_*}\left(\nabla_*\cdot\boldsymbol{Q}_{o11} - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}Q_{211} - B_{xx}P^*\right) \\
&\quad \left. +\int_0^{t_*}\frac{\tau_c^2}{4}\exp\left(\frac{-2(t_*-\tau)}{\tau_c}\right)\frac{\partial^2}{\partial\tau^2}\left(\nabla_*\cdot\boldsymbol{Q}_{o11} - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}Q_{211} - B_{xx}P^*\right)d\tau\right] \\
&= -\left[\frac{\tau_c}{2}\left(\nabla_*\cdot\boldsymbol{Q}_{o11} - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}Q_{211} - B_{xx}P^*\right)\right. \\
&\quad \left. -\frac{\tau_c^2}{4}\frac{\partial}{\partial t_*}\left(\nabla_*\cdot\boldsymbol{Q}_{o11} - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}Q_{211} - B_{xx}P^*\right) + ...\right]. \qquad \text{(D.1)}
\end{aligned}
$$

D.2 Integration of Q_{122}

$$
\begin{aligned}
Q_{122} &= -\int_0^{t_*} \exp\left(\frac{-3(t_*-\tau)}{\tau_c}\right)\left(\nabla_*\cdot\boldsymbol{R}_{o122} - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)d\tau \\
&= -\left[\frac{\tau_c}{3}\left(\nabla_*\cdot\boldsymbol{R}_{o122} - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)\right. \\
&\quad \left. -\int_0^{t_*}\frac{\tau_c}{3}\exp\left(\frac{-3(t_*-\tau)}{\tau_c}\right)\frac{\partial}{\partial\tau}\left(\nabla_*\cdot\boldsymbol{R}_{o122} - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)d\tau\right] \\
&= -\left[\frac{\tau_c}{3}\left(\nabla_*\cdot\boldsymbol{R}_{o122} - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)\right. \\
&\quad -\frac{\tau_c^2}{9}\frac{\partial}{\partial t_*}\left(\nabla_*\cdot\boldsymbol{R}_{o122} - \frac{t_*}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right) \\
&\quad \left. +\int_0^{t_*}\frac{\tau_c^2}{9}\exp\left(\frac{-3(t_*-\tau)}{\tau_c}\right)\frac{\partial^2}{\partial\tau^2}\left(\nabla_*\cdot\boldsymbol{R}_{o122} - \frac{\tau}{\tau_f}\frac{\partial}{\partial x_*}\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)d\tau\right] \\
&= -\left[\frac{\tau_c}{3}\left(\nabla\cdot\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)\right. \\
&\quad -\frac{\tau_c^2}{9}\frac{\partial}{\partial t}\left(\nabla\cdot\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right) + \frac{\tau_c^2}{9}U_x^\infty\frac{\partial}{\partial x}\left(\nabla\cdot\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right) \\
&\quad \left. +\int_0^{t}\frac{\tau_c^2}{9}\exp\left(\frac{-3(t-\tau)}{\tau_c}\right)\frac{\partial^2}{\partial\tau^2}\left(\nabla\cdot\boldsymbol{R}_{o122} - B_{yy}j_x - 2B_{xy}j_y\right)d\tau\right]. \qquad \text{(D.2)}
\end{aligned}
$$

D.3 Derivation of the R terms

$$R_{1111} = \frac{6}{4}\tau_c B_{xx} P_{x1} = \frac{6}{4}\frac{1}{\tau_c} 2 D_{xx} D_{xx} \frac{1}{\tau_c} P^* = \frac{3}{\tau_c^2} D_{xx} D_{xx} P^*, \tag{D.3}$$

$$\begin{aligned} R_{1112} &= \frac{3}{4}\tau_c B_{xx} P_{x2} + \frac{3}{4}\tau_c B_{xy} P_{x1} = \frac{3}{4}\frac{1}{\tau_c} 2 D_{xx} \frac{D_{xy}}{\tau_c} P^* + \frac{3}{4}\frac{1}{\tau_c} 2 D_{xy} D_{xx} \frac{1}{\tau_c} P^* \\ &= \frac{3}{\tau_c^2} D_{xx} D_{xy} P^*, \end{aligned} \tag{D.4}$$

$$R_{1113} = \frac{3}{4}\tau_c B_{xx} P_{x3} = 0, \tag{D.5}$$

$$\begin{aligned} R_{1222} &= \frac{3}{4}\tau_c B_{xy} P_{y2} + \frac{3}{4}\tau_c B_{yy} P_{x2} = \frac{3}{4}\frac{1}{\tau_c} 2 D_{xy} D_{yy} \frac{1}{\tau_c} P^* + \frac{3}{4}\frac{1}{\tau_c} 2 D_{yy} \frac{D_{xy}}{\tau_c} P^* \\ &= \frac{3}{\tau_c^2} D_{xy} D_{yy} P^*, \end{aligned} \tag{D.6}$$

$$\begin{aligned} R_{1122} &= \frac{1}{4}\tau_c B_{xx} P_{y2} + \frac{1}{4}\tau_c B_{yy} P_{x1} + \tau_c B_{xy} P_{x2} \\ &= \frac{1}{4}\frac{1}{\tau_c} 2 D_{xx} D_{yy} \frac{1}{\tau_c} P^* + \frac{1}{4}\frac{1}{\tau_c} 2 D_{yy} D_{xx} \frac{1}{\tau_c} P^* + \frac{1}{\tau_c} 2 D_{xy} \frac{D_{xy}}{\tau_c} P^* \\ &= \frac{1}{\tau_c^2} D_{xx} D_{yy} P^* + \frac{2}{\tau_c^2} D_{xy} D_{xy} P^*, \end{aligned} \tag{D.7}$$

$$\begin{aligned} R_{1133} &= \frac{1}{4}\tau_c B_{xx} P_{z3} + \frac{1}{4}\tau_c B_{zz} P_{x1} = \frac{1}{4}\frac{1}{\tau_c} 2 D_{xx} \frac{D_{zz}}{\tau_c} P^* + \frac{1}{4}\frac{1}{\tau_c} 2 D_{zz} D_{xx} \frac{1}{\tau_c} P^* \\ &= \frac{1}{\tau_c^2} D_{xx} D_{zz} P^*, \end{aligned} \tag{D.8}$$

$$R_{1123} = \frac{1}{4}\tau_c B_{xx} P_{y3} + \frac{1}{2}\tau_c B_{xy} P_{x3} = 0, \tag{D.9}$$

$$R_{1223} = \frac{1}{2}\tau_c B_{xy} P_{y3} + \frac{1}{4}\tau_c B_{yy} P_{x3} = 0, \tag{D.10}$$

$$\begin{aligned} R_{1233} &= \frac{1}{4}\tau_c B_{xy} P_{z3} + \frac{1}{4}\tau_c B_{zz} P_{x2} = \frac{1}{4}\frac{1}{\tau_c} 2 D_{xy} \frac{D_{zz}}{\tau_c} P^* + \frac{1}{4}\frac{1}{\tau_c} 2 D_{zz} \frac{D_{xy}}{\tau_c} P^* \\ &= \frac{1}{\tau_c^2} D_{xy} D_{zz} P^*, \end{aligned} \tag{D.11}$$

$$R_{2222} = \frac{6}{4}\tau_c B_{yy} P_{y2} = \frac{6}{4}\frac{1}{\tau_c} 2 D_{yy} D_{yy} \frac{1}{\tau_c} P^* = \frac{3}{\tau_c^2} D_{yy} D_{yy} P^*, \tag{D.12}$$

$$R_{3333} = \frac{6}{4}\tau_c B_{zz} P_{z3} = \frac{6}{4}\frac{1}{\tau_c} 2 D_{zz} \frac{D_{zz}}{\tau_c} P^* = \frac{3}{\tau_c^2} D_{zz} D_{zz} P^*, \tag{D.13}$$

$$R_{1333} = \frac{3}{4}\tau_c B_{zz} P_{x3} = 0, \tag{D.14}$$

$$\begin{aligned} R_{2233} &= \frac{1}{4}\tau_c B_{yy} P_{z3} + \frac{1}{4}\tau_c B_{zz} P_{y2} = \frac{1}{4}\frac{1}{\tau_c} 2 D_{yy} \frac{D_{zz}}{\tau_c} P^* + \frac{1}{4}\frac{1}{\tau_c} 2 D_{zz} D_{yy} \frac{1}{\tau_c} P^* \\ &= \frac{1}{\tau_c^2} D_{yy} D_{zz} P^*, \end{aligned} \tag{D.15}$$

$$R_{2223} = \frac{3}{4}\tau_c B_{yy} P_{y3} = 0, \tag{D.16}$$

$$R_{2333} = \frac{3}{4}\tau_c B_{zz} P_{y3} = 0. \tag{D.17}$$

D.4 Derivation of the Q terms

$$
\begin{aligned}
Q_{113} &= \frac{\tau_c}{3}B_{xx}j_z - \frac{\tau_c}{3}\frac{\partial R_{1113}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2113}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3113}}{\partial z} \\
&= -\frac{2}{3\tau_c}D_{xx}D_{zz}\frac{\partial P^*}{\partial z} - 0 - 0 - \frac{\tau_c}{3}\frac{1}{\tau_c^2}D_{xx}D_{zz}\frac{\partial P^*}{\partial z} \\
&= -\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial P^*}{\partial z}, \qquad \text{(D.18)} \\
Q_{222} &= \tau_c B_{yy}j_y - \frac{\tau_c}{3}\frac{\partial R_{1222}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2222}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3222}}{\partial z} \\
&= \frac{2}{\tau_c}D_{yy}\left(-D_{xy}\frac{\partial P^*}{\partial x} - D_{yy}\frac{\partial P^*}{\partial y}\right) - \frac{\tau_c}{3}\frac{3}{\tau_c^2}D_{xy}D_{yy}\frac{\partial P^*}{\partial x} - \frac{\tau_c}{3}\frac{3}{\tau_c^2}D_{yy}D_{yy}\frac{\partial P^*}{\partial y} - 0 \\
&= -\frac{3}{\tau_c}D_{yy}D_{xy}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{yy}D_{yy}\frac{\partial P^*}{\partial y}, \qquad \text{(D.19)} \\
Q_{123} &= \frac{\tau_c}{3}B_{xy}j_z - \frac{\tau_c}{3}\frac{\partial R_{1123}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2123}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3123}}{\partial z} \\
&= -\frac{2}{3\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial z} - 0 - 0 - \frac{\tau_c}{3}\frac{1}{\tau_c^2}2D_{xy}D_{zz}\frac{\partial P^*}{\partial z} \\
&= -\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial z}, \qquad \text{(D.20)} \\
Q_{111} &= \tau_c B_{xx}j_x - \frac{\tau_c}{3}\frac{\partial R_{1111}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2111}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3111}}{\partial z} \\
&= \frac{2}{\tau_c}D_{xx}\left(-D_{xx}\frac{\partial P^*}{\partial x} - D_{xy}\frac{\partial P^*}{\partial y}\right) - \frac{\tau_c}{3}\frac{3}{\tau_c^2}D_{xx}D_{xx}\frac{\partial P^*}{\partial x} - \frac{\tau_c}{3}\frac{3}{\tau_c^2}D_{xx}D_{xy}\frac{\partial P^*}{\partial y} - 0 \\
&= -\frac{3}{\tau_c}D_{xx}D_{xx}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial y}, \qquad \text{(D.21)} \\
Q_{112} &= \frac{\tau_c}{3}B_{xx}j_y + \frac{2\tau_c}{3}B_{xy}j_x - \frac{\tau_c}{3}\frac{\partial R_{1112}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2112}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3112}}{\partial z} \\
&= \frac{2}{3\tau_c}D_{xx}\left(-D_{xy}\frac{\partial P^*}{\partial x} - D_{yy}\frac{\partial P^*}{\partial y}\right) + \frac{4}{3\tau_c}D_{xy}\left(-D_{xx}\frac{\partial P^*}{\partial x} - D_{xy}\frac{\partial P^*}{\partial y}\right) \\
&\quad - \frac{\tau_c}{3}\frac{3}{\tau_c^2}D_{xx}D_{xy}\frac{\partial P^*}{\partial x} - \frac{\tau_c}{3}\left(\frac{1}{\tau_c^2}D_{xx}D_{yy} + \frac{2}{\tau_c^2}D_{xy}D_{xy}\right)\frac{\partial P^*}{\partial y} - 0 \\
&= -\frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial P^*}{\partial y} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial P^*}{\partial y}, \qquad \text{(D.22)} \\
Q_{233} &= \frac{\tau_c}{3}B_{zz}j_y - \frac{\tau_c}{3}\frac{\partial R_{1233}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2233}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3233}}{\partial z} \\
&= \frac{2}{3\tau_c}D_{zz}\left(-D_{xy}\frac{\partial P^*}{\partial x} - D_{yy}\frac{\partial P^*}{\partial y}\right) - \frac{\tau_c}{3}\frac{1}{\tau_c^2}D_{xy}D_{zz}\frac{\partial P^*}{\partial x} - \frac{\tau_c}{3}\frac{1}{\tau_c^2}D_{yy}D_{zz}\frac{\partial P^*}{\partial y} - 0 \\
&= -\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{yy}D_{zz}\frac{\partial P^*}{\partial y}, \qquad \text{(D.23)}
\end{aligned}
$$

$$
\begin{aligned}
Q_{333} &= \tau_c B_{zz} j_z - \frac{\tau_c}{3}\frac{\partial R_{1333}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2333}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3333}}{\partial z} \\
&= \frac{2}{\tau_c} D_{zz}\left(-D_{zz}\frac{\partial P^*}{\partial z}\right) - 0 - 0 - \frac{\tau_c}{3}\frac{3}{\tau_c^2} D_{zz} D_{zz}\frac{\partial P^*}{\partial z} \\
&= -\frac{3}{\tau_c} D_{zz} D_{zz}\frac{\partial P^*}{\partial z}, \qquad \text{(D.24)} \\
Q_{133} &= \frac{\tau_c}{3} B_{zz} j_x - \frac{\tau_c}{3}\frac{\partial R_{1133}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2133}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3133}}{\partial z} \\
&= \frac{2}{3\tau_c} D_{zz}\left(-D_{xx}\frac{\partial P^*}{\partial x} - D_{xy}\frac{\partial P^*}{\partial y}\right) - \frac{\tau_c}{3}\frac{1}{\tau_c^2} D_{xx} D_{zz}\frac{\partial P^*}{\partial x} - \frac{\tau_c}{3}\frac{1}{\tau_c^2} D_{xy} D_{zz}\frac{\partial P^*}{\partial y} - 0 \\
&= -\frac{1}{\tau_c} D_{xx} D_{zz}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c} D_{xy} D_{zz}\frac{\partial P^*}{\partial y}, \qquad \text{(D.25)} \\
Q_{223} &= \frac{\tau_c}{3} B_{yy} j_z - \frac{\tau_c}{3}\frac{\partial R_{1223}}{\partial x} - \frac{\tau_c}{3}\frac{\partial R_{2223}}{\partial y} - \frac{\tau_c}{3}\frac{\partial R_{3223}}{\partial z} \\
&= -\frac{2}{3\tau_c} D_{yy} D_{zz}\frac{\partial P^*}{\partial z} - 0 - 0 - \frac{\tau_c}{3}\frac{1}{\tau_c^2} D_{yy} D_{zz}\frac{\partial P^*}{\partial z} \\
&= -\frac{1}{\tau_c} D_{yy} D_{zz}\frac{\partial P^*}{\partial z}. \qquad \text{(D.26)}
\end{aligned}
$$

D.5 Derivation of the P terms

Inserting the Q_{111}, Q_{211} and Q_{311} (setting o to x, y, z) into the equation for P_{x1} yields:

$$\begin{aligned}
P_{x1} = &-\frac{\tau_c}{2}\left(\frac{\partial}{\partial x}Q_{111} + \frac{\partial}{\partial y}Q_{211} + \frac{\partial}{\partial z}Q_{311} - B_{xx}P^*\right) \\
&+ \frac{\tau_c^2}{4}\left(\frac{\partial}{\partial t}\left(\frac{\partial}{\partial x}Q_{111} + \frac{\partial}{\partial y}Q_{211} + \frac{\partial}{\partial z}Q_{311} - B_{xx}P^*\right)\right. \\
&\left.+U_x^\infty\frac{\partial}{\partial x}\left(\frac{\partial}{\partial x}Q_{111} + \frac{\partial}{\partial y}Q_{211} + \frac{\partial}{\partial z}Q_{311} - B_{xx}P^*\right)\right) - ... \\
= &-\frac{\tau_c}{2}\left(\frac{\partial}{\partial x}\left(-\frac{3}{\tau_c}D_{xx}D_{xx}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial y}\right)\right. \\
&+ \frac{\partial}{\partial y}\left(-\frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial P^*}{\partial y} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial P^*}{\partial y}\right) \\
&\left.+\frac{\partial}{\partial z}\left(-\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial P^*}{\partial z}\right) - B_{xx}P^*\right) \\
&+ \frac{\tau_c^2}{4}\left(\frac{\partial}{\partial t}\left(\frac{\partial}{\partial x}Q_{111} + \frac{\partial}{\partial y}Q_{211} + \frac{\partial}{\partial z}Q_{311} - B_{xx}P^*\right)\right. \\
&+ U_x^\infty\frac{\partial}{\partial x}\left(\frac{\partial}{\partial x}\left(-\frac{3}{\tau_c}D_{xx}D_{xx}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial y}\right)\right. \\
&+ \frac{\partial}{\partial y}\left(-\frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial P^*}{\partial y} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial P^*}{\partial y}\right) \\
&\left.\left.+\frac{\partial}{\partial z}\left(-\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial P^*}{\partial z}\right) - B_{xx}P^*\right)\right) - ...
\end{aligned} \tag{D.27}$$

The only remaining terms are the time derivative of P^* and the U_x^∞ component of P^* from the last terms.

$$
\begin{aligned}
P_{x1} = &-\frac{\tau_c}{2}\left(-\frac{3}{\tau_c}D_{xx}D_{xx}\frac{\partial^2 P^*}{\partial x^2} - \frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y}\right.\\
&-\frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} - \frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial y^2} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial y^2}\\
&\left.-\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial^2 P^*}{\partial z^2} - \frac{2}{\tau_c^2}D_{xx}P^*\right)\\
&+\frac{\tau_c^2}{4}\left(-\frac{\partial P^*}{\partial t}\frac{2}{\tau_c^2}D_{xx} - U_x^\infty\frac{\partial P^*}{\partial x}\frac{2}{\tau_c^2}D_{xx}\right)\\
= &-\frac{\tau_c}{2}\left(-\frac{3}{\tau_c}D_{xx}D_{xx}\frac{\partial^2 P^*}{\partial x^2} - \frac{6}{\tau_c}D_{xx}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y}\right.\\
&-\frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial y^2} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial y^2}\\
&\left.-\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial^2 P^*}{\partial z^2} - \frac{2}{\tau_c^2}D_{xx}P^*\right)\\
&+\frac{1}{2}D_{xx}\left(U_x^\infty\frac{\partial P^*}{\partial x} + \frac{\partial j_x}{\partial x} + \frac{\partial j_y}{\partial y} + \frac{\partial j_z}{\partial z} - U_x^\infty\frac{\partial P^*}{\partial x}\right)\\
= &-\frac{\tau_c}{2}\left(-\frac{3}{\tau_c}D_{xx}D_{xx}\frac{\partial^2 P^*}{\partial x^2} - \frac{6}{\tau_c}D_{xx}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y}\right.\\
&-\frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial y^2} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial y^2}\\
&\left.-\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial^2 P^*}{\partial z^2} - \frac{2}{\tau_c^2}D_{xx}P^*\right)\\
&-\frac{1}{2}D_{xx}\left(D_{xx}\frac{\partial^2 P^*}{\partial x^2} + D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{yy}\frac{\partial^2 P^*}{\partial y^2} + D_{zz}\frac{\partial^2 P^*}{\partial z^2}\right)\\
= &\frac{1}{\tau_c}D_{xx}P^* + D_{xx}D_{xx}\frac{\partial^2 P^*}{\partial x^2} + 2D_{xx}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial y^2}.
\end{aligned}
\tag{D.28}
$$

The other components of $\boldsymbol{P}$ read:

$$
\begin{aligned}
P_{x2} &= -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o12} - B_{xy}P^*\right) - \frac{\tau_c^2}{4}\left(B_{xy}\frac{\partial P^*}{\partial t} + U_x^\infty B_{xy}\frac{\partial P^*}{\partial x}\right)\\
&= -\frac{\tau_c}{2}\left(\frac{\partial}{\partial x}\left(-\frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial P^*}{\partial y} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial P^*}{\partial y}\right)\right.\\
&\quad + \frac{\partial}{\partial y}\left(-\frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial P^*}{\partial x} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{xy}D_{yy}\frac{\partial P^*}{\partial y}\right)\\
&\quad \left.+\frac{\partial}{\partial z}\left(-\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial z}\right) - \frac{2}{\tau_c^2}D_{xy}P^*\right) - \frac{\tau_c^2}{4}\left(\frac{2}{\tau_c^2}D_{xy}\frac{\partial P^*}{\partial t} + U_x^\infty\frac{2}{\tau_c^2}D_{xy}\frac{\partial P^*}{\partial x}\right)\\
&= -\frac{\tau_c}{2}\left(-\frac{3}{\tau_c}D_{xx}D_{xy}\frac{\partial^2 P^*}{\partial x^2} - \frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial x\partial y} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y}\right.\\
&\quad - \frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial x\partial y} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} - \frac{3}{\tau_c}D_{xy}D_{yy}\frac{\partial^2 P^*}{\partial y^2}\\
&\quad \left.-\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial^2 P^*}{\partial z^2} - \frac{2}{\tau_c^2}D_{xy}P^*\right) - \frac{1}{2}\left(D_{xy}\frac{\partial P^*}{\partial t} + U_x^\infty D_{xy}\frac{\partial P^*}{\partial x}\right)\\
&= \frac{1}{2}\left(3D_{xx}D_{xy}\frac{\partial^2 P^*}{\partial x^2} + D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial x\partial y} + 2D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y}\right.\\
&\quad + D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial x\partial y} + 2D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + 3D_{xy}D_{yy}\frac{\partial^2 P^*}{\partial y^2}\\
&\quad \left.+D_{xy}D_{zz}\frac{\partial^2 P^*}{\partial z^2} + \frac{2}{\tau_c}D_{xy}P^*\right)\\
&\quad - \frac{1}{2}D_{xy}\left(D_{xx}\frac{\partial^2 P^*}{\partial x^2} + D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{yy}\frac{\partial^2 P^*}{\partial y^2} + D_{zz}\frac{\partial^2 P^*}{\partial z^2}\right)\\
&= \frac{1}{\tau_c}D_{xy}P^* + D_{xx}D_{xy}\frac{\partial^2 P^*}{\partial x^2} + D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{xy}D_{yy}\frac{\partial^2 P^*}{\partial y^2},
\end{aligned}
\tag{D.29}
$$

$$
\begin{aligned}
P_{x3} &= -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o13}\right)\\
&= -\frac{\tau_c}{2}\left(\frac{\partial}{\partial x}\left(-\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial P^*}{\partial z}\right) + \frac{\partial}{\partial y}\left(-\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial z}\right)\right.\\
&\quad \left.+\frac{\partial}{\partial z}\left(-\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial y}\right)\right)\\
&= D_{xx}D_{zz}\frac{\partial^2 P^*}{\partial x\partial z} + D_{xy}D_{zz}\frac{\partial^2 P^*}{\partial y\partial z}.
\end{aligned}
\tag{D.30}
$$

The other components follow subsequently.

$$
\begin{aligned}
P_{y2} &= -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o22} - B_{yy}P^*\right) - \frac{\tau_c^2}{4}\left(B_{yy}\frac{\partial P^*}{\partial t} + U_x^\infty B_{yy}\frac{\partial P^*}{\partial x}\right)\\
&= \frac{1}{\tau_c}D_{yy}P^* - \frac{\tau_c}{2}\left(\frac{\partial}{\partial x}\left(-\frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial P^*}{\partial x} - \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{xy}D_{yy}\frac{\partial P^*}{\partial y}\right)\right.\\
&\quad + \frac{\partial}{\partial y}\left(-\frac{3}{\tau_c}D_{yy}D_{xy}\frac{\partial P^*}{\partial x} - \frac{3}{\tau_c}D_{yy}D_{yy}\frac{\partial P^*}{\partial y}\right) + \frac{\partial}{\partial z}\left(-\frac{1}{\tau_c}D_{yy}D_{zz}\frac{\partial P^*}{\partial z}\right)\Bigg)\\
&\quad - \frac{1}{2}\left(D_{yy}\frac{\partial P^*}{\partial t} + U_x^\infty D_{yy}\frac{\partial P^*}{\partial x}\right)\\
&= \frac{1}{\tau_c}D_{yy}P^* + \frac{\tau_c}{2}\left(\frac{1}{\tau_c}D_{xx}D_{yy}\frac{\partial^2 P^*}{\partial x^2} + \frac{2}{\tau_c}D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial x^2} + \frac{3}{\tau_c}D_{xy}D_{yy}\frac{\partial^2 P^*}{\partial x\partial y}\right.\\
&\quad + \frac{3}{\tau_c}D_{yy}D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + \frac{3}{\tau_c}D_{yy}D_{yy}\frac{\partial^2 P^*}{\partial y^2} + \frac{1}{\tau_c}D_{yy}D_{zz}\frac{\partial^2 P^*}{\partial z^2}\Bigg)\\
&\quad - \frac{1}{2}D_{yy}\left(D_{xx}\frac{\partial^2 P^*}{\partial x^2} + D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{yy}\frac{\partial^2 P^*}{\partial y^2} + D_{zz}\frac{\partial^2 P^*}{\partial z^2}\right)\\
&= \frac{1}{\tau_c}D_{yy}P^* + D_{xy}D_{xy}\frac{\partial^2 P^*}{\partial x^2} + 2D_{xy}D_{yy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{yy}D_{yy}\frac{\partial^2 P^*}{\partial y^2}, \qquad \text{(D.31)}\\
P_{y3} &= -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o23}\right)\\
&= -\frac{\tau_c}{2}\left(\frac{\partial}{\partial x}\left(-\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial z}\right) + \frac{\partial}{\partial y}\left(-\frac{1}{\tau_c}D_{yy}D_{zz}\frac{\partial P^*}{\partial z}\right)\right.\\
&\quad + \frac{\partial}{\partial z}\left(-\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{yy}D_{zz}\frac{\partial P^*}{\partial y}\right)\Bigg)\\
&= D_{xy}D_{zz}\frac{\partial^2 P^*}{\partial x\partial z} + D_{yy}D_{zz}\frac{\partial^2 P^*}{\partial y\partial z}, \qquad \text{(D.32)}\\
P_{z3} &= -\frac{\tau_c}{2}\left(\nabla\cdot\boldsymbol{Q}_{o33} - B_{zz}P^*\right) - \frac{\tau_c^2}{4}\left(B_{zz}\frac{\partial P^*}{\partial t} + U_x^\infty B_{zz}\frac{\partial P^*}{\partial x}\right)\\
&= \frac{D_{zz}}{\tau_c}P^* - \frac{\tau_c}{2}\left(\frac{\partial}{\partial x}\left(-\frac{1}{\tau_c}D_{xx}D_{zz}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial y}\right)\right.\\
&\quad + \frac{\partial}{\partial y}\left(-\frac{1}{\tau_c}D_{xy}D_{zz}\frac{\partial P^*}{\partial x} - \frac{1}{\tau_c}D_{yy}D_{zz}\frac{\partial P^*}{\partial y}\right) + \frac{\partial}{\partial z}\left(-\frac{3}{\tau_c}D_{zz}D_{zz}\frac{\partial P^*}{\partial z}\right)\Bigg)\\
&\quad - \frac{1}{2}D_{zz}\left(\frac{\partial P^*}{\partial t} + U_x^\infty D_{zz}\frac{\partial P^*}{\partial x}\right)\\
&= \frac{D_{zz}}{\tau_c}P^* + \frac{1}{2}\left(D_{xx}D_{zz}\frac{\partial^2 P^*}{\partial x^2} + D_{xy}D_{zz}\frac{\partial^2 P^*}{\partial x\partial y}\right.\\
&\quad + D_{xy}D_{zz}\frac{\partial^2 P^*}{\partial x\partial y} + D_{yy}D_{zz}\frac{\partial^2 P^*}{\partial y^2} + 3D_{zz}D_{zz}\frac{\partial^2 P^*}{\partial z^2}\Bigg)\\
&\quad - \frac{1}{2}D_{zz}\left(D_{xx}\frac{\partial^2 P^*}{\partial x^2} + D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{xy}\frac{\partial^2 P^*}{\partial x\partial y} + D_{yy}\frac{\partial^2 P^*}{\partial y^2} + D_{zz}\frac{\partial^2 P^*}{\partial z^2}\right)\\
&= \frac{D_{zz}}{\tau_c}P^* + D_{zz}D_{zz}\frac{\partial^2 P^*}{\partial z^2}. \qquad \text{(D.33)}
\end{aligned}
$$